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A COMPARATIVE STUDY OF COLD PLASMA GENERATION AT ATMOSPHERIC PRESSURE IN VARIOUS WORKING MEDIA

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INTRODUCTION

A **plasma display** panel (PDP) is a type of flat panel display common to large TV displays 30 inches (76 cm) or larger. They are called "plasma" displays because the technology utilizes small cells containing electrically charged ionized gases, or what are in essence chambers more commonly known as fluorescent lamps.

Plasmas are conductive assemblies of charged particles, neutrals and fields that exhibit collective effects. Further, plasmas carry electrical currents and generate magnetic fields. Plasmas are the most common form of matter, comprising more than 99% of the visible universe, and permeate the solar system, interstellar and intergalactic environments.

Plasmas are radically multi scale in two senses

(1) Most plasma systems involve electro-dynamic coupling across micro-, meso- and macroscale.

(2) Plasma systems occur over most of the physically possible ranges in space, energy and density scales. The figure here illustrates where many plasma systems occur in terms of typical densities and temperatures.

However, the full range of possible plasma density, energy (temperature) and spatial scales go far beyond this illustration. For example, some space plasmas have been measured to be lower in density than 10 to the power -10 per cubic meter or (10exp-10)/m3 - 13 orders of magnitude less than the scale shown in the figure! On the other extreme, quark-gluon plasmas (although mediated via the strong force field versus the electromagnetic field) are extremely dense nuclear states of matter. For temperature (or energy), some plasma crystal states produced in the laboratory have temperatures close to absolute zero. In contrast, space plasmas have been measured with thermal temperatures above 10+9 degrees Kelvin and cosmic rays (a type of plasma with very large gyroradii) are observed at energies well above those produced in any man-made accelerator laboratory. Considering Powers of 10 is useful for grasping the unique way in which plasmas are radically multi-scale in space, energy and density.

Because plasmas are conductive and respond to electric and magnetic fields and can be efficient sources of radiation, they can be used in innumerable applications where such control is needed or when special sources of energy or radiation are required.

In physics and chemistry, **plasma** is a state of matter similar to gas in which a certain portion of the particles are ionized. Heating a gas may ionize (reduce the number of electrons in) its molecules or atoms, thus turning it into a plasma, which contains charged particles: positive ions and negative electrons. Ionization can be induced by other means, such as strong electromagnetic field applied with a laser or microwave generator, and is accompanied by the dissociation of molecular bonds, if present.

The presence of a non-negligible number of charge carriers makes the plasma electrically conductive so that it responds strongly to electromagnetic fields. Plasma, therefore, has properties quite unlike those of solids, liquids, or gases and is considered a distinct state of matter. Like gas, plasma does not have a definite shape or a definite volume unless enclosed in a container; unlike gas, under the influence of a magnetic field, it may form structures such as filaments, beams and double layer. Some common plasmas are stars and neon signs. In the universe, plasma is the most common state of matter for ordinary matter, most of which is in the rarefied intergalactic plasma (particularly intracluster medium) and in stars.

Plasma is loosely described as an electrically neutral medium of positive and negative particles (i.e. the overall charge of plasma is roughly zero). It is important to note that although they are unbound, these particles are not 'free'. When the charges move they generate electrical currents with magnetic fields, and as a result, they are affected by each other's fields. This governs their collective behavior with many degrees of freedom. A definition can have three criteria:

1. **The plasma approximation**: Charged particles must be close enough together that each particle influences many nearby charged particles, rather than just interacting with the closest particle (these collective effects are a distinguishing feature of a plasma). The plasma approximation is valid when the number of charge carriers within the sphere of influence (called the *Debye sphere* whose radius is the Debye screening length) of a particular particle is higher than unity to provide collective behavior of the charged particles. The average number of particles in the Debye sphere is given by the plasma parameter, "Λ" (the Greek letter Lambda).

2. **Bulk interactions**: The Debye screening length (defined above) is short compared to the physical size of the plasma. This criterion means that interactions in the bulk of the plasma are more important than those at its edges, where boundary effects may take place. When this criterion is satisfied, the plasma is Quasineutrality.

3. **Plasma frequency**: The electron plasma frequency (measuring plasma oscillations of the electrons) is large compared to the electron-neutral collision frequency (measuring frequency

of collisions between electrons and neutral particles). When this condition is valid, electrostatic interactions dominate over the processes of ordinary gas kinetics.

4. **Magnetization:** Plasma with a magnetic field strong enough to influence the motion of the charged particles is said to be magnetized. A common quantitative criterion is that a particle on average completes at least one gyration around the magnetic field before making a collision, i.e., $\omega_{ce}/\nu_{coll} > 1$, where ω_{ce} is the "electron gyro frequency" and ν_{coll} is the "electron collision rate". It is often the case that the electrons are magnetized while the ions are not. Magnetized plasmas are *anisotropic*, meaning that their properties in the direction parallel to the magnetic field are different from those perpendicular to it. While electric fields in plasmas are usually small due to the high conductivity, the electric field associated with a plasma moving in a magnetic field is given by $E = -v \times B$ (where E is the electric field, v is the velocity, and B is the magnetic field),and is not affected by Debye shielding.

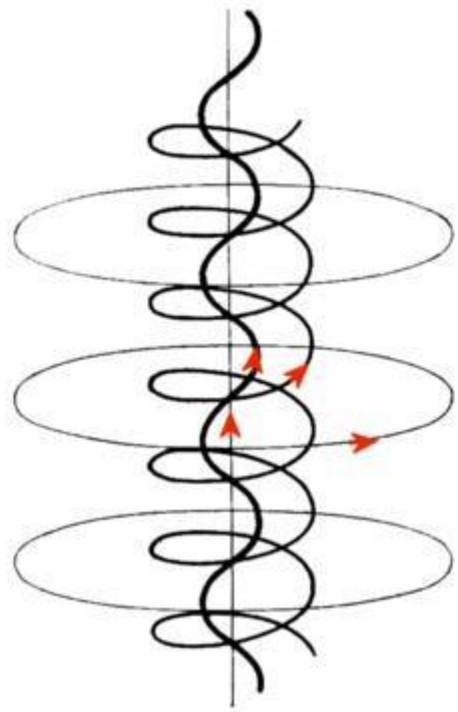
SHOCKS OR DOUBLE LAYERS

Plasma properties change rapidly (within a few Debye lengths) across a two-dimensional sheet in the presence of a (moving) shock or (stationary) double layer. Double layers involve localized charge separation, which causes a large potential difference across the layer, but does not generate an electric field outside the layer. Double layers separate adjacent plasma regions with different physical characteristics, and are often found in current carrying plasmas. They accelerate both ions and electrons.

ELECTRIC FIELDS AND CIRCUITS

Quasineutrality of a plasma requires that plasma currents close on themselves in electric circuits. Such circuits follow Kirchhoff's circuit laws and possess a resistance and inductance. These circuits must generally be treated as a strongly coupled system, with the behavior in each plasma region dependent on the entire circuit. It is this strong coupling between system elements, together with nonlinearity, which may lead to complex behavior. Electrical circuits in plasmas store inductive (magnetic) energy, and should the circuit be disrupted, for example, by a plasma instability, the inductive energy will be released as plasma heating and acceleration. This is a common explanation for the heating that takes place in the solar corona. Electric currents, and in particular, magnetic-field-aligned electric currents (which are sometimes generically referred to as "Baekeland"), are also observed in the Earth's aurora, and in plasma filaments.

Mathematical descriptions



The complex self-constricting magnetic field lines and current paths in a field-aligned Baekeland current that can develop in a plasma.

To completely describe the state of a plasma, we would need to write down all the particle locations and velocities and describe the electromagnetic field in the plasma region. However, it is generally not practical or necessary to keep track of all the particles in a plasma.

ARTIFICIAL PLASMAS

Most artificial plasmas are generated by the application of electric and/or magnetic fields. Plasma generated in a laboratory setting and for industrial use can be generally categorized by:

- The type of power source used to generate the plasma—DC, RF and microwave
- The pressure they operate at—vacuum pressure (< 10 mTorr or 1 Pa), moderate pressure (~1 Torr or 100 Pa), atmospheric pressure (760 Torr or 100 kPa)
- The degree of ionization within the plasma—fully, partially, or weakly ionized
- The temperature relationships within the plasma—thermal plasma ($T_e = T_{ion} = T_{gas}$), non-thermalor "cold" plasma ($T_e >> T_{ion} = T_{gas}$)

- The electrode configuration used to generate the plasma
- The magnetization of the particles within the plasma—magnetized (both ion and electrons are trapped in Larmor orbits by the magnetic field), partially magnetized (the electrons but not the ions are trapped by the magnetic field), non-magnetized (the magnetic field is too weak to trap the particles in orbits but may generate Lorentz forces)
- The application

In contrast, non-thermal plasmas are capable of operating effectively at low temperature. Electrons of sufficient energy colliding with the background gas can result in a low level of dissociation, excitation and ionization without an appreciable increase in the gas enthalpy. This is the realm of cold plasmas in which the electron temperature can exceed the temperature of the heavy particles by several orders of magnitude. Because the ions and the neutrals remain relatively cold, this characteristic provides the possibility of using these plasmas for low temperature plasma chemistry and for the lighting industry. In such cases, the plasma chemistry is driven by the electrons, which cause ionization, molecular excitation and production of radicals. The advantage of low bulk gas temperature is exploited when the surroundings of the plasma cell are required to be protected from heat and when the kinetics of the processes need to be controlled. The huge field of lighting and display industry [Marshak, 1984; Waymouth, 1991; Dakin, 1993; Lister *et al.*, 2004; Zissis and Rouffet, 2006] relies heavily on the cold plasma technology, as most of the optical radiation sources in the world use non-thermal plasma on a large scale for the generation of radiant optical energy which is one of the most abundant forms of energy available to the mankind. Our world cannot be conceived without an optical light source. Most of the light that we live by-outdoors as well as indoors, both during the day and at night–comes from the plasmas, the electrically charged gas that results when numerous atoms are broken down into electrically charged particles. Outdoors during the day the light comes from the sun, which is nothing but the plasma; at night along with some sunlight reflected by the moon and some star light, there are fluorescent lamps and high intensity arc discharge lamps to light the way which are again plasma based. Thus, both during the day and night, most of the light comes from the plasma only. Light is also crucial for other areas of our lives. It is important for plant growth and for the liberation of oxygen by photosynthesis, the sun's plasma thus being responsible for much of our food and for the very air that we breathe. All in all, optical light system is an important socio-economic factor and its development is an integral part of any sustainable development and of any program involving improvement of the quality of life.

Human genius has created a large number of its own optical radiation sources for generating optical energy. These optical radiation sources, besides serving the basic purpose of providing light are also very vital in a great variety of scientific, industrial, technological and medical applications and in several other diversified fields of science and technology. Cathode ray tubes emit impulses that activatescreens of computer monitors and television. X rays are used not only as a diagnostic tool in medicine, but also as an analytical tool in the inspection of manufactured products and other composite structures.Microwaves are used not only in cooking or as a means of heating rubber or plastic but also in a wide variety of communication applications. Infrared radiations are used in heating, in analytical chemistry and in electronics.

During the last two decades, in addition to the radiations in the above mentioned regions of the electromagnetic spectrum, the field of ultraviolet radiation has gained a major impetus in diverse areas of science and technology. Although ultraviolet (UV) radiations have been in use for decades in areas including the medical field, photochemistry, photobiology, microelectronics, and the analytical fields; but more recently these radiations have found applications in advanced areas such as photon induced material processing, Very Large Scale Integration, communication and information technology and industries based on electronics and photonic technologies which often require ultraviolet optical radiation sources [Boyd *et al.*, 2003; Griesbeck *et al.*, 2003; Kogelschatz, 2004].

UV optical sources [Rice, 1997; Malik *et al.*, 2001; Laroussi *et al.*, 2002] not only play an important role in the scientific and industrial circles, but also have a tremendous impact on our society. In the 21st century, the contribution of UV optical sources to other fields has become essential to address growing global environmental problem. In the field of environmental technology, UV optical sources are widely utilized for ozone generation, for elimination of pollutants in air (chlorofluorocarbons, dioxins, etc.), treatment of drinking water, and also in wastewater treatment.

Also, photon induced material processing [Esrom and Kogelschatz, 1992; Bergonzo and Boyd, 1993; Esrom *et al.*, 2000; Kogelschatz *et al.*, 2000] using ultraviolet optical sources offers an unprecedented potential. These sources have been used in several areas including surface modification, material deposition/coating of metals, dielectrics (high and low dielectric constant materials), and semi conducting layers, hardening of paints, lacquers, and adhesives, for printing and lamination, in automotive and equipment engineering.

UV radiation sources also have a potential impact on textile and polymer technology [Ersom *et al.*, 1992; Mehnert *et al.*, 2002] where they are used in surface treatment, e.g. surface modification of polymers, dry etching of polymers, synthesis of hydrophilic polymers to increase adhesion between metal and polymer, surface cleaning and surface etching including three-dimensional applications, and textile finishing. Furthermore, they are used in several photon initiated scientific and industrial applications [Lomaev *et al.*, 2006a] such as in the field of photo-chemistry, e.g. for photo-chlorination, photo-sulpho-oxidation, photo-nitrosylization, photo-oxidation, photo mineralization, actinometry etc., in photo-medicine, such as for the treatment of skin conditions, tanning etc., in photobiology for photo inactivation, photo regulation and photo destruction.

Keeping in view the enormous potential applications of ultraviolet optical sources, in brief, it can be deduced that the development of ultraviolet optical sources is a task of great significance. In order to carry out photon initiated scientific and industrial applications, large area ultraviolet optical radiation sources are required. Therefore, during the last few decades, considerable efforts have been extended by technologists and engineers to the generation of energetic VUV and UV photons, which represent radiations in the UV region of the electromagnetic spectrum. A great deal of work has now been focused on the designing and development of several optical sources, major ones being the non-thermalplasma based excimer optical sources. In the present work also, the primary focus of research will be onthe study of design issues of cold plasma based VUV and UV sources. But, before proceeding further towards defining the objectives of this research work, it is necessary to have a fundamental understanding of the UV spectrum, its classification and also a comparative study of the various sources of UV generation.

OBJECTIVES

Keeping in view the growing importance of non-thermal plasma based excimer sources; the presentwork has been planned with the following objectives:

- To study possible application of non-thermal plasma sources for excimer generation and toinvestigate miscellaneous process parameters to devise a DBD reactor.
- To investigate electrical equivalent model of the DBD and to determine the discharge excitation parameters of excimer generation scheme.
- Implementation of the electrical equivalent network model in Matlab Simulink tool to studythe electrical behavior of the DBD reactor.
- To study the frequency response of DBD, and consideration of load matching conditions ofDBD plasma load.

LITERATURE REVIEW

Electrical discharges have been known to the mankind for more than a century, beginning well before the discovery of electrons. Until the discovery of the transistor and the development of integrated circuits, the use of ionized gas was essential for the control of electric current within the electrical power and communication industries, creating the interdisciplinary field known as gaseous electronics. Although most of the gaseous components within electronic systems disappeared with the development of semiconductor technology, but still the manufacture of semiconductor devices is dependent upon the use of gas discharge through plasma processing technique. Moreover, understanding of ionized gas is still essential to the lighting and display industries, despite the advent of light emitting diode, solid statelasers and liquid crystal displays.

Although the application of an electrical discharge in plasma chemical reactions has a long history, but the synthesis of excimer using DBD technology is a relatively new field. One of the most important early empirical landmarks in the history of DBD electrical discharges was the invention of silent discharge by Werner Von Siemens in 1857. Siemens proposed a novel type of electrical gas discharge that could generate ozone from atmospheric oxygen or air. This was achieved by subjecting a flow of oxygen or air to the influence of a dielectric-barrier discharge maintained in a narrow annular gap between two coaxial glass tubes by an alternating electric field of sufficient amplitude. Since theelectric current was forced to pass through the glass walls acting as dielectric barriers the discharge was referred to as the dielectric-barrier discharge. Some oxygen molecules in the air flowing through the discharge gap between the glass tubes were converted to ozone. The novel feature of this discharge apparatus was that the electrodes were positioned outside the discharge chamber and were not in contact with the plasma.

Dielectric barrier discharge technology, a technique for producing atmospheric non-thermal plasma, provides effective means for efficient ionization of the gas at atmospheric pressure that is well suited for several chemical plasma processes. The classical dielectric barrier discharge utilizes planar or cylindrical electrode arrangements with at least one dielectric layer placed between the electrodes. There is a wide variance in the terminology used for the discharge in literature. The names assigned to the phenomena include silent discharge, barrier discharge, ozonizer discharge, dielectric barrier glow discharge and filamentary dielectric barrier discharge.

All the studies conducted on DBD in context of an excimer source during the past years may be dividedinto four major categories: studies related to micro discharge formation and determination of their properties; numerical modeling and studies of chemical kinetics in different optical working media; electric circuit analysis, modeling, simulation of DBD load and power source design; and finally the various applications of DBD.

A few years after Siemens original invention, DBD technology was investigated for ozone generationat industrial level. Using this technology large scale ozone production for the treatment of drinking water started in Europe during 1900. The initial efforts were oriented towards the industrial developments of ozone generator. During the development of the ozonizer, efforts were made to investigate electrical breakdown in barrier discharge. In 1940, Loeb and Meek had independently presented their concepts of electric breakdown mechanism of discharge initiation at high pressure in theform of the streamer breakdown mechanism [Loeb and Meek, 1940]. In 1943, Manley proposed a novelmethod for determining the dissipated power in DBDs by using closed voltage charge Lissajous figures and derived the famous power formula for DBD which relates the dissipated power to the operating frequency, the applied peak voltage and some other parameters of the barrier discharge [Manley, 1943].

The term excimer (excited dimmer) [Stevens and Hutton, 1960] was initially proposed by Stevens and Hutton in 1960. The acronym excimer was basically applied to the unstable molecular complexes formed from an atom in the ground state and another one in an electronically excited state. Although Basov and coworkers first demonstrated an excimer laser [Basov *et al.*, 1970] and later, several workers [Rhodes, 1974; Rhodes, 1979; Lakoba and Yakovlenko, 1980; Smirnov, 1983] carried out investigations on the properties of an excimer molecule on a large scale. But, it was Tanka [Tanka, 1955] who first demonstrated that rare gas excimer can be formed in electrical gas discharge excited by fast pulses, microwaves or silent discharge. The second excimer continua of rare gas had also been obtained by Tanka in a much simpler configuration used as spectroscopic light source for absorption measurement in vacuum ultraviolet spectral range. These devices could be regarded as the first excimer UV optical sources, although they were not sealed at that time. Further work on spectroscopic light sources using rare gas excimer emission was continued by Soviet researchers [Volkova *et al.*, 1984]. Investigations were extended to obtain excimer emission from rare gas halides [Shevera *et al.*, 1980] and mercury halides [Malinin *et al.*, 1980]. In all these early investigations, the radiation was extracted through relatively small windows in the discharge vessels.

Even until about ten years ago, ozone generation was the major industrial application of DBD technology with thousands of installed ozone generating facilities. Extensive research activities on DBD using modern diagnostic and modeling tools led to a better understanding of the plasma physical and chemical processes in ozonizer discharge. These research efforts not only led to improved ozone generators, but also resulted into a number of additional applications of DBD. The first application of DBD as ozonizer is still one of the most important application but, in the meantime, other fields of applications such as surface modification, plasma chemical vapor deposition, pollution control, excitation of CO_2 lasers, excimer UV generation, and several others have also started being investigated. The DBDs are now widely used for several different industrial and technological applications.

Based on the experience with industrial ozonizers, the turning point in the field of DBD research was the possibility of using this technology for exciting rare gases using silent discharge, a special non equilibrium discharge that can be operated at atmospheric pressure. Discharge modeling conducted on DBD to identify optimum discharge parameters with respect to gap width, dielectric properties, operating pressure, voltage waveform and driving frequency [Eliasson *et al.*, 1987; Kogelschatz, 1988] have shown that DBD technology has a vital role in excimer formation. Initial investigations on excimer lamp were based on kinetic models developed for excimer lasers. Starting in 1988, first models specifically made to simulate excimer formation in discharge lamps started appearing in the literature.

The first major attempt on excimer formation utilizing the DBD technology was made by Kogelschatz and his coworkers in 1988 [Eliasson and Kogelschatz, 1988; Eliasson *et al.*, 1988; Kogelschatz, 1990; Gellert and Kogelschatz, 1991; Kogelschatz, 1992]. They realized that the excimer known from electron beam experiment can also be formed in silent discharges, if at least one of the electrodes is made transparent to the excimer formation. It was shown that a large number of different excimers can be generated in a simple gas discharge with a DBD. The effect of geometry, different gas mixtures on excimer formation, possibility of obtaining different wavelengths, physical parameters of micro discharge and reaction kinetics of excimer formation in DBD have also been investigated. It is possible to excite excimer radiation in VUV/UV or even in the visible range. Spectroscopic investigations were performed with these sources. Considering the large number of known excimers, one is led to believe that an efficient radiation source for any desired spectral range can be developed. Model calculations describing electrical breakdown and micro discharge formation were also presented [Eliasson and Kogelschatz, 1991; Eliasson and Kogelschatz, 1994].

A few years after the idea of Eliasson and Kogelschatz, a large number of excimer spectra were recorded in such discharge configurations. Eliasson and Gellert in 1990 investigated excimer formation in a mixture of mercury and rare gases excited by dielectric barrier discharge. A theoretical model which considers both the discharge formation and the charge particle kinetics of HgXe was presented [Eliasson and Gellert, 1990]. Besides rare gases, excited complexes also known as exciplexes have been obtained in barrier discharge. The predominant reaction path leading to the exciplex formation is the harpooning reaction. Since F atom attacks silica surface ∗ KrF , ∗ ArF and 2 F lamps have only been used in through flow system in the laboratory [Kumagai and Obara, 1989a; Kumagai and Obara, 1989b].

The application of DBD technology for pumping excimer formation gained importance, and another major contribution in the field of excimer synthesis utilizing DBD technology was made by Boyd and his co-workers at the University College, London. They conducted studies on the chemical kinetics of different excimers using different sets of conditions in barrier discharge and also investigated diverse applications of excimer UV source. Powerful and efficient ∗ XeBr and ∗ XeI exciplexes have been successfully tested in the laboratory [Zhang and Boyd, 1996; Zhang and Boyd, 1998; Boyd and Zhang, 2000; Boyd and Zhang, 2001].

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visible wavelengths. Most of the ordinary (or baryonic) matter in the universe, however, is found in the intergalactic medium, which is also a plasma, but much hotter, so that it radiates primarily as X-rays. The current scientific consensus is that about 96% of the total energy density in the universe is not plasma or any other form of ordinary matter, but a combination of cold dark matter and dark energy.

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