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## Low Frequency Glow Discharge Sources as an Impressive Technique for Different Applications: A mini review article

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### ABSTRACT

Low-Frequency (LF) glow discharge has been used for different applications such as plasma chemical vapor deposition (PCVD) of thin films, medical treatment applications such as disinfection, wound healing and industry of recombinant proteins. LF glow discharge source is regard as an inexpensive simple construction, easy to operate and low consumption power compared with High-Frequency (HF) and Direct current (DC) glow discharge sources. Highlight of recent advances in using LF glow discharge source in previous applications are demonstrated in this article. In the first section, the working principles of DC, HF and LF gas discharge are covered, along with a comparison of the three techniques. The advancements of LF gas discharge in thin film deposition PCVD and in medical treatment are discussed in the following section. Finally, a variety of computer simulation models of LF-PECVD technique and Cold atmospheric plasma CAP, which can be generated by LF, are presented for the previous applications.

### INTRODUCTION

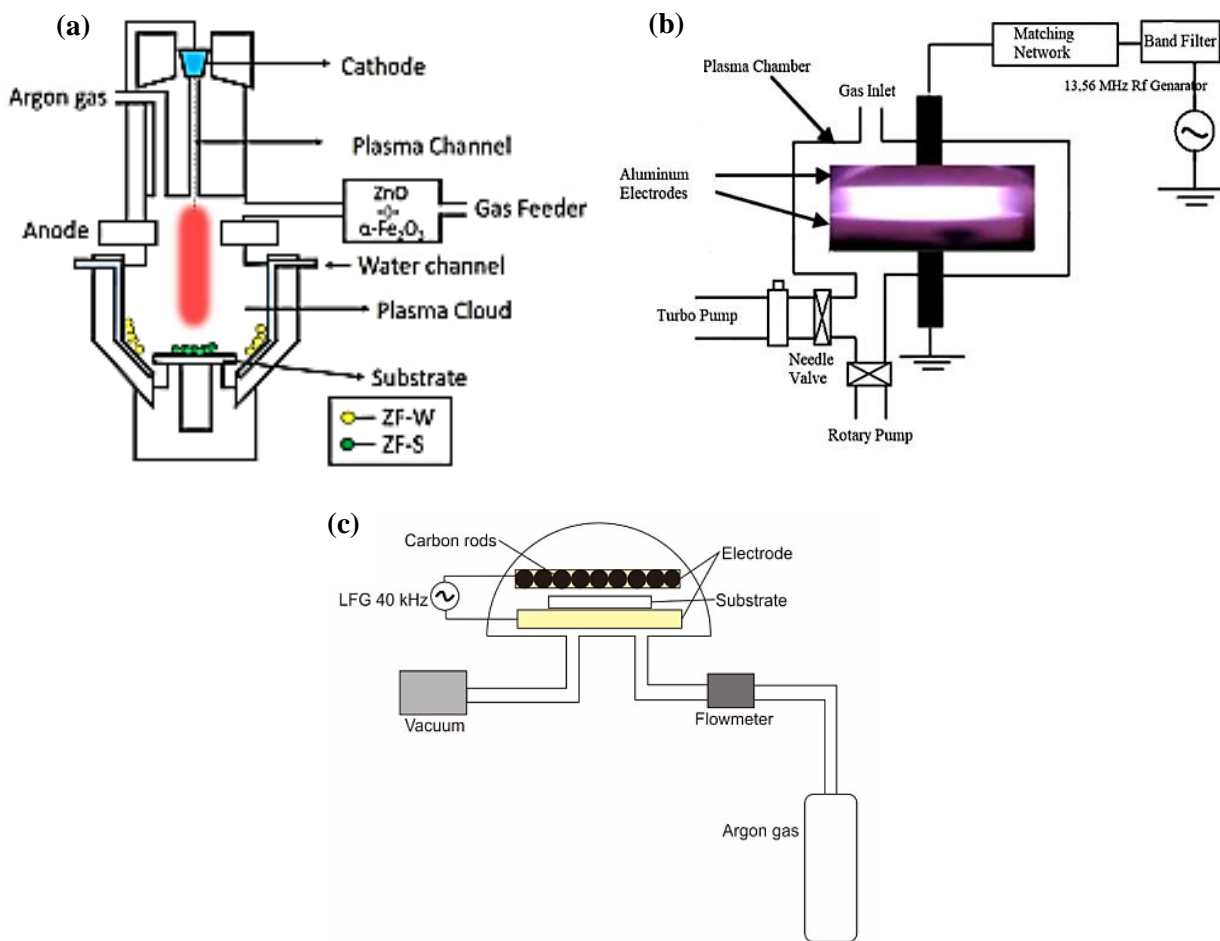
Materials plays important role in technological advances in electronics, aerospace, vehicle industry and preparation of thin material film and generation of submicron spherical particles. With increasing demand for high performance materials attention has been given to new processing techniques such as gas discharge and plasma for material fabrication and treatment. Direct current and arc discharge spry technique are the most standard technologies for such application [1-2]. Radio frequency gas discharge is used for spraying and coating of metal surfaces. [3-5].

On the other hand treatment of some diseased skin, disinfection of (Bacteria/Fungi/Viruses), wound healing, biofilms treatment and the industry of recombinant proteins by plasma become a subject of interest [6-7]. Highlights of some of these advances by radio frequency (RF) gas discharge which is divided into HF and LF gas discharge are presented in this article. The first section deals briefly with DC, HF and LF gas discharge techniques. Second section presents the advances in PCVD of thin films by LF gas discharge then it followed by advances in Cold

atmospheric plasma (CAP) which can be easily generated by LF gas discharge. The final section demonstrates computer modeling for PCVD of thin films deposition process by LF gas discharge and CAP technique.

### DC, RF and LF glow discharge sources for PCVD of thin films

A schematic diagram showing the basic differences between DC, HF and LF gas discharge techniques is given in Fig. 1. In a DC gas discharge, a high voltage is applied between two electrodes (anode and cathode). A direct current flows through the gas in the gap between the two electrodes in a vacuum chamber and the gas is ionizing and forming plasma. In this process, gaseous ions are ejected into the source material, causing gas molecules, atoms, and ions to sputter off the target surface. This method makes controlling the sputtering and coating process easy. DC sputtering technique has difficulties with insulating materials due to high operating cost, low deposition rates degradation of some materials, introduction of impurities and irregular deposition. Fig. 1(a) shows the schematic diagram of DC gas discharge source [8].



**Fig. (1): (a) DC gas discharge source [8] (b) RF –gas discharge source (Capacitive type) [9] and (c) LF- gas discharge source (Capacitive type) [10]**

Radio frequency (RF) gas discharge is divided into high frequency (HF) range (from ten to hundreds of MHz) and low frequency range (LF) which frequency is less than 10 MHz. Both are called RF power sources. [9]

In radiofrequency (RF) gas discharge, the plasma is formed by radiofrequency induction utilizing an RF power source and an oscillator frequency 1 MHz or higher, typically operates at frequencies between 5 and 30 MHz and power range from 50 to 400 kW. RF power source is connected to the cathode within the vacuum chamber causing the cathode's electric polarity to alternate. In the other half-cycle, target atoms spewed by positive ions striking the target are deposited on the substrate and form a layer. Consequently, the electrons reach the target when it holds the positive pole in the half-cycle and neutralize the positive ions gathered on the target surface. RF sputtering technique is regard more efficient compared to DC gas discharge source but at a higher cost. Fig. 1(b)

shows the schematic diagram of RF gas discharge source (Capacitive type) [9].

In Low frequency (LF) gas discharge, the plasma generation is similar to radiofrequency (RF) gas discharge, and the lower frequency of LF gas discharge is less oscillatory than the operating frequency range of RF gas discharge. Fig. 1(c) shows the schematic diagram of LF gas discharge source (Capacitive type) [10].

LF excitation is not “DC-like”, the plasma persists throughout the RF cycle — but the frequency is much lower than (1/sheath transit time). The entire energy associated with the RF voltage will thus be acquired by ions that enter the sheath close to the wafers, minus any losses from scattering with neutral molecules or atoms. The sheath voltage is small when the electrode is the anode, but large when it is the cathode and because of the mechanisms that support the plasma, electrode voltages at LF range are often substantially larger than those at RF

range. LF plasma depends on secondary emission at surfaces: that is, an ion strikes the surface and generates one or more secondary electrons. The sheath voltage accelerates the electrons into the plasma and repels them from the cathode. The electrons have enough energy to ionize molecules when they get to the plasma region, and the voltage rises until the ions lost at the sheath borders are made up for by this secondary ionization. On the other hand for RF excitation, Sheath edge motion is rapid. This has the benefit of forcing a large amount of capacitive current to pass through the plasma in order to alternately charge and discharge the sheaths. With a lower sheath voltage, the displacement current can aid in ionizing molecules, sustaining the plasma. Naturally, the ion density must match the electron density in the plasma's bulk, but the resulting electron density in a high-frequency plasma is usually much higher than that of a low-frequency plasma. Furthermore, as the sheath transit time is now usually greater than one RF cycle, the ion energy striking the surfaces tends to be rather consistent in time rather than cycling between very intense and very gentle ions [11]. The majority of the RF and LF method's explanation can be found in the references [12-13] .

**The advantage of LF gas discharge is as follows:**

- LF gas discharge generation can spread across the vacuum chamber and is not restricted to the cathode or target surface, plasma can be sustained in less working gas pressure (1–15 mTorr), which reduces the amount of atoms that sputter into chamber molecules and increases the mean free path for target atoms extend within the vacuum chamber [14 - 16].
- By eliminating charge build up on the cathode surface, a more homogenous layer can be deposited.
- Target lifetime is increased with sputtering because a larger target surface is involved in the sputtering process, which reduces "race track erosion" on the target's surface.
- low cost compared to DC and RF discharge sources.

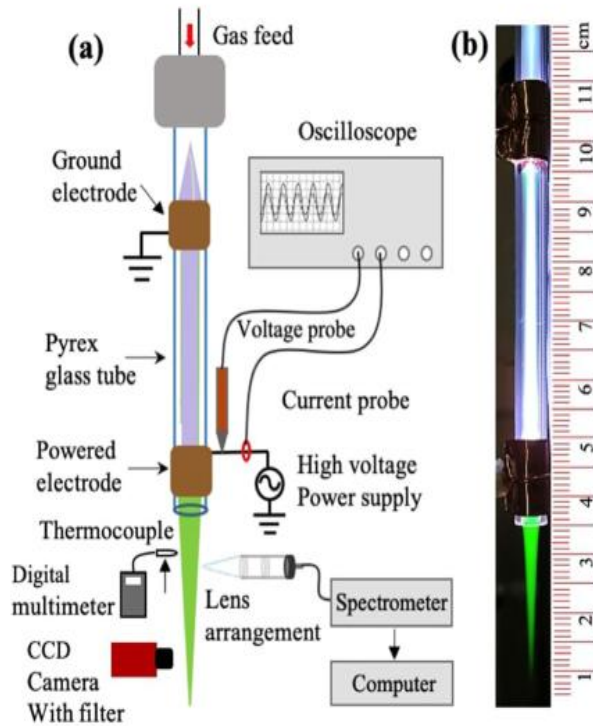
Carbon Nano Walls (CNWs) synthesized by PECVD technology is regard as one of the recent developments which uses gas discharge for thin film deposition process. Zhang et al. have shown fast growth rates and unique morphological structures, opening up new application possibilities for which based on gas discharge technique [17].

Different methods have been applied with different parameters, such as PECVD and DC gas discharge,

producing different morphologies and characteristics. Graphite was evaporated using a DC arc-discharge method in a vacuum chamber supplied with hydrogen by Ando et al. producing smaller nanometric petal-like structures and interwoven petal-like sheets [18]. By depositing vertical graphene sheets atop SiOx nanowire networks. Wang et al. created three-dimensional graphene networks with lots of sharp edge locations [19]. The morphology and nanostructure of carbon nanowalls (CNWs) generated by PECVD were used to explore the electrochemical activity of CNWs [20]

In addition to nanostructure applications, the LF gas discharge technique has medical applications as well. In medical application, Cold atmospheric plasma (CAP) terminology is used. CAP can be generated efficiently and at a reasonable cost by using LF gas discharge. Fig. 2 shows the Schematic diagram of the experimental setup of the LF gas discharge for CAP-jet technique. The plasma in CAP definition is an infinite system consisting of many free electrons, ions, and neutral particles, with macroscopic time and space scales. It can be generated by glow discharge, corona discharge, or arc discharge at normal atmospheric pressure. CAP plasma jet are used in dermatological applications for reduction of itch after treatment for one minute over a period of 30 days and no side effects have been observed [21] . Another case study of CAP for local infection control, a patient with a chronic postoperative ear infection in another case study experienced a highly substantial reduction in pain [22] .

In Disinfection (Bacteria/Fungi/Viruses), CAP treatment for two- minute has been demonstrated to be beneficial for wound healing against a range of bacteria, including significant skin and wound pathogens, treatment a fungal infection of the nail and preventing of viruses [22-25]. For biofilm treatment applications, contaminated biofilms have been treated by plasma techniques [26]. Dental implants can provide unique anatomical challenges when it comes to removing dental plaque. This emphasizes the need for a therapy that can be provided with CAP that has a consistent cleaning efficacy and an effective penetration depth to kill bacteria in thick biofilm [27]. Increased Thrombotic thrombocytopenic purpura (TTP) metabolism at every antimicrobial concentration employed showed that the combination of CAP plasma and antibiotics was efficient at changing the metabolic activity of the biofilms, indicating greater susceptibility against the three strains of *P. aeruginosa* studied [28]. *P. aeruginosa* is a gram-negative, aerobic, non-spore forming rod and has negative effect on tissues.

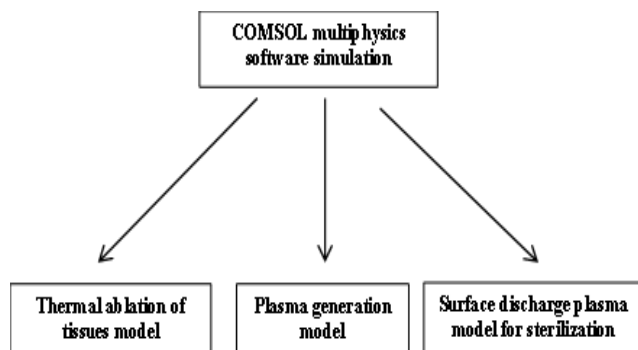


**Fig. (2): (a) Schematic diagram of the experimental setup LF gas discharge for CAP technique (b) Photograph of the system [S. Jaiswal et. al]**

### Computer simulation for PCVD

Most computer simulations studies in this area have been devoted to computation of plasma temperature, density and particle flow energy and effect of previous parameters on thin film deposition.

A coupled multiphysics field model built on the COMSOL Multi-physics platform is formed to simulate  $\text{SiN}_x\text{:H}$  thin film deposition via LF-PECVD technique by J. Zhou [29]. COMSOL multiphysics is a software used to simulate designs and processes in the field of engineering and scientific research. It used in many applications areas such as CAP sources.



**Fig. (3): COMSOL multiphysics software in simulation of medical and biological applications [27-30]**

Figure 3 shows the ability of COMSOL multiphysics software in simulation of CAP in medical and biological applications [30-33]. Combining analysis for the flow field, heat field, chemical reaction field, and plasma field simulates the deposition process. The results show that while pressure and temperature are the main process parameters that affect film uniformity, temperature has the greatest impact on the rate of deposition. Two dimensional simulation of Argon discharge was performed. Ion density and electric field, the ion flow bombarding the substrate, and the ion energy at the substrate was displayed by X. Xu [34]. The frequency effect of the LF source is investigated. With the low frequency, the uniformities of the ion density and ion flow distributions are almost independent. However, the increase in low frequency dramatically reduces the uniformities of ion density and ion flux distributions when two sources are coupled. Additionally, it is discovered that while the homogeneity of ion bombardment energy reaching the substrate varies somewhat in the radial direction, the amplitude of ion bombarding energy reduces significantly with an increase in low frequency. It is also noted that the ion density in the radial direction is non-uniform and an increase in LF voltage reduces the homogeneity of the ions density distribution and radial ion flux distribution. By changing the amplitude of the LF voltage, it caused worse uniformity. LF pulsed direct current planar magnetron (PDCM), which is widely used in reactive sputtering to produce thin films, was explored by S. B. Q. Tran [35]. A fluid model in a two-dimensional axisymmetric P-DCM setup was used. The results demonstrated that the argon ion flows to both, the electrical potentials change during the pulse cycle, and the magnet traps electrons close to the target. Target and substrate resulting in deposition flow. Pulsing leads in higher electron density and temperature but lower deposition rate as compared to non-pulsed DCM. As the pulse frequency increases, the electron energy increases but the electron density, argon flux on the target, and deposition rate all decrease. on the other hand, Increasing duty cycle, results in a decrease in electron energy and density as well as an increase in argon flux on target and deposition rate and the duty cycle and frequency have an inverse relationship with time-averaged electron density and time-averaged discharge voltage magnitude, respectively. Another simulation study was done by Y. Zhou [36] to show the LF effect on plasma density and ion energy/angular distribution. Distribution in low pressure (2 Pa) capacitively coupled

DF plasma Based on a combination of experimental and kinetic particle simulations methods. Using the electrostatic PIC code in conjunction with Monte Carlo processing of collision processes (PIC/MCCs), fundamental physics is revealed by capturing electron and ion dynamics. According to the results, as the frequency decreases from 6.8 MHz to 40 kHz, plasma density undergoes a moderate decline initially, followed by an increase, reaching a maximum at 400 kHz. The greater emission of electron-induced secondary electrons and the mitigation of the LF source's modulation influence on high-frequency electron heating are jointly responsible for the improved plasma density. Additionally, ion energy with a narrower angular spread can be obtained with a lower frequency.

### Computer simulation for CAP in medical applications

Most computer simulations in this area have been devoted to simulate the fluid dynamic and plasma parameters on the biological cells and tissues.

COMSOL software was utilized to simulate the gas discharge and fluid dynamics, utilizing the governing equations as described by Nguyen, Tam [37]. Two dimensional- FEM simulation studies were carried out to compare the plasma parameters between ignitions by DC-pulsed or sinusoidal power supplies. Additionally, the OES spectrum was used to calculate the electron density and temperature ( $T_e$ ) in the plasma jet's AC mode. OD600 which equivalent is equivalent to approximately  $3 \times 10^7$  cells/mL and Capacity measurements revealed that the treatment of the AC power source had no effect on the yeast cell, proving the safety of the AC plasma jet for yeast cells. The results demonstrate that there are no appreciable changes to cell growth or viability. Both AC pulsed DC plasma configurations produced more protein, with the pulsed DC plasma producing higher protein levels. The results demonstrated a rise in protein synthesis that is reliant on the plasma treatment dose, which is equal to the treatment's duration.

In the plasma jet ignited by AC and pulsed DC, the increase was found to be as high as 45% and 33%, respectively. This suggests that while pulsed DC and AC power supply can both encourage yeast cells to create more protein, the AC setup has demonstrated a little more productive result. The results that have been provided imply that gene expression is positively impacted by the atmospheric pulsed plasma and that cell viability and growth are not affected by plasma exposure and the enzymatic activity has increased by plasma induction. These results show that atmospheric pulsed plasma is a

reliable and scalable instrument in the recombinant protein industry [38].

Electric field mathematical modeling for cold atmospheric pressure plasma-jet is done by P. Vafeas [39]. In this study, an electric field analytical solution for plasma jet reactor configurations has been developed. The first plasma source consists of a dielectric tube with a single outer electrode electrically biased. The second consists of an insulating tube with two outer electrodes, one electrically biased and the other grounded. The motivation was to obtain analytical solutions for the electric field that could be used in plasma simulations, rather than time-consuming numerical solutions. The single electrode case results have demonstrated the critical importance that the location of the zero-potential boundary condition plays in providing an accurate electric field calculation. This work presented an analytical solution that is beneficial for estimating the electric field in plasma simulations because it is hard to impose the specific boundary condition at infinity in numerical solutions. Furthermore, the results for the double-electrode reactor demonstrated that, depending on the electrode configuration, the presence of a grounded electrode may not have an impact on the electric field and, in turn, the conditions for the plasma's initiation. The modeling from this study could be a useful tool in designing of cold atmospheric pressure plasma-jet for tailored applications.

A Changes in the permeability of cell membranes to various reactive oxygen species (ROS) ( $O_2$ ,  $HO_2$ ,  $OH$ , and  $H_2O_2$ ) during plasma therapy and natural conditions were investigated using Molecular dynamic (MD) simulation[40]. In this study, GROningen MOlecular Simulation (GROMOS) was used to study the relationship between ROS density and distribution. It was investigated how various ROS species behaved at the water-membrane contact as well as how the membrane's permeability and shape changed at the microscopic level. The oxidation rate was calculated and the microscopic penetration and distribution of various ROS in the cell membrane were characterized. It also covered the presence of phospholipids and cholesterol. The results demonstrated that the cholesterol oxidation varies significantly depending on the kind of ROS and phospholipids present. The ROS produced by the plasma also has an impact on how the phospholipids are arranged in the cell membrane. The distribution of ROS at the membrane-water interface is influenced by the molar concentrations of phospholipid molecules thereby affecting the efficiency of plasma destruction of the cell membranes. In natural conditions, hydrophobic  $O_2$  is prefer to enter the interior of the cell,



while a strong barrier by the lipid bilayer is formed to hydrophilic ROS (OH, HO<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>). Among them, H<sub>2</sub>O<sub>2</sub> has the highest barrier energy due to hydrogen bonding. Under plasma treatment, the energy barrier of HO<sub>2</sub> decreases significantly, while the other types of ROS do not change greatly.

## CONCLUSION

From the above, it can be seen that the ability of LF gas discharge techniques to synthesis high quality small size particle fine structure and deposit thin film is possible and reliable. Also CAP technique which can be generated by LF gas discharge can be applied to achieve promising and low cost solutions in medical applications such as treatment of biological tissues and it gains the tissues immune properties against disease and infection. It will need ongoing research in this extremely active area of LF gas discharge therapy to fully comprehend the mechanisms of action of CAP and to properly explore its therapeutic potential. In addition to this field, plasma in this way will also open the way to other application fields. In addition to this field, CAP technique by LF gas discharge in this way will also open the way to other application fields.

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