



Sterilization of Microorganisms Contaminated Surfaces and its Treatment with Dielectric Barrier Discharge Plasma

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Abstract

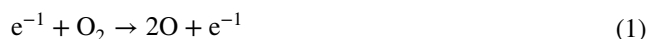
Dielectric barrier discharge (DBD) is a promising method of producing non-thermal plasma, which is widely used in variety of industrial and biological applications including disinfection/sterilization. Plasma sterilization offers a faster, less toxic and versatile alternative to conventional sterilization techniques. 45 kV, 50 kHz high voltage high-frequency power supply was designed for generating DBD plasma. Experimental studies were conducted using DBD plasma on growth control in algae, breakdown of complex phenols for chemical wastewater treatment and generation of UV, ozone and other reactive species using DBD plasma discharges in ambient air. A portable DBD plasma-based sterilization system is developed for fighting the Covid-19 pandemic. Quartz tube is used as a dielectric medium between copper foil—SS electrodes and 17 kV, 30 kHz pulse is applied across it, which produces intense DBD across SS mesh. This system can sterilize and disinfect the microorganism contaminated surfaces, garments and used disposable protective gears with UV, ozone and short-lived molecules of metastable states and excited chemical species of nitrogen and oxygen produced during the DBD plasma discharges in ambient air.

Keywords Plasma · Dielectric barrier · Sterilization · Power supply · Spectroscopy

Introduction

Sterilization is a process that completely eliminates or destructs all living microorganism from the contaminated surfaces. Autoclaving, dry heat, ethylene oxide fumigation, radioactive irradiation are the conventional methods of sterilization/disinfection. The major drawbacks of conventional techniques are: heat sensitive, requires longer time exposure, usage of chemicals. The ideal sterile should provide short time, low temperature, versatility of operation and harmless for the operator and the surfaces (Moisan et al. 2001). Plasma sterilization has simplicity in its operation, non-radio isotopic technique, requires very less time, and provides all the advantages for biological applications (Laroussi 2005; Gallagher et al. 2007; Kim et al. 2015). Dielectric barrier discharge (DBD) produces many reactive species during the plasma discharge in air. In ambient air, nitrogen and oxygen

molecules are dissociated by the energetic electrons and ions in the discharge mostly leading to the formation of radicals (O and OH), ozone (O₃), Nitrogen Oxides (NO, NO₂) and excited N₂ through several reaction pathways. When compressed air is passed through the plasma zone, various reactions are initiated. The free energetic electrons interact with the compressed air, they dissociate oxygen molecules as



Following this, ozone is formed by three-body collision reaction:



M is collision partner. It can be O₂, O₃ or N₂.

DBD plasma treatment is very promising method and the results suggest that free radicals generated by discharge plasma played an important role in sterilization and disinfection (Tanion et al. 2005; Xia et al. 2019). Using a relatively small, low temperature, atmospheric, dielectric barrier discharge surface plasma generator, ≥ 6 log reduction in the concentration of vegetative bacterial and yeast cells within 4 min and ≥ 6 log reduction of *Geobacillus stearothermophilus* spores within 20 min was achieved

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(Mastanaiah et al. 2013). In this article, we will discuss the mechanism for generating DBD plasma with high-frequency power supply and its applications for growth control in algae, treatment of chemical wastewater and production of metastable states and excited chemical reactive species of nitrogen and oxygen in air for sterilization of contaminated surfaces.

High Voltage and High Frequency Power Supply

DBD has at least one insulating barrier between the electrodes so the DC current cannot pass through it and requires ac voltages. The dielectric constant of insulating barrier, thickness and the frequency of voltage determine the displacement current that can pass through it. To initiate the discharge in the gap the electric field should be high between the dielectric medium and electrodes (Kogelschatz et al. 1997). The power supply requires operation over varying plasma load condition so series resonance converter circuit is preferred as it has simple circuit topology, capacity to withstand short circuit and provides constant current characteristics over wider range of load conditions (Surender and Anurag 2016). A 45 kV, 50 kHz high voltage high-frequency power supply was designed and constructed for generating DBD plasma (Surender and Anurag 2015). The specifications of the high voltage pulse power supply are mentioned in Table 1. The power supply requires 230 V ac at 50 Hz and produces high voltages pulses of 45 kV at 50 kHz maximum. The output voltage can be varied from 1 to 45 kV and the pulse frequency can also be varied from 1 to 50 kHz. The rise time and the fall time of the high voltage pulses are 2 μ s and the pulse width is also 2 μ s. The protection circuit is included in the power supply for over current, under voltage, and over temperature. The open-circuit pulse voltage waveform is shown in Fig. 1.

Table 1 Specification of high voltage power supply

Parameter	Value
Input voltage (V)	230 V AC, 50 Hz
Output voltage (kV)	1–45
Maximum output current (Amp)	0.5
Frequency (kHz)	1–50
Peak power (kW)	22.5
Average power (kW)	1.2
Pulse rise time (μ s)	2
Pulse fall time (μ s)	2
Pulse width (μ s)	2

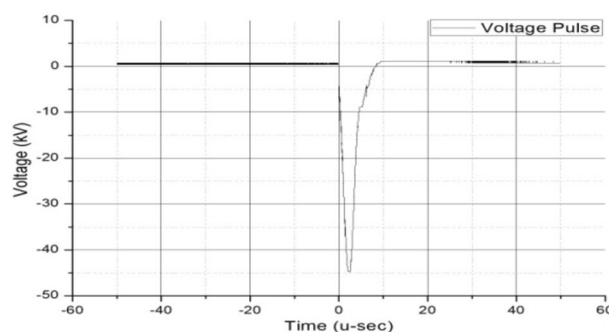


Fig. 1 Open circuit voltage of power supply

Algae Growth Control

The undesirable growth of algal biomass both at surface water and industrial water storages cause nuisance in maintaining the water quality parameters and reutilization of the water. Therefore, dielectric barrier discharge plasma treatment was applied to the algae samples as an advanced oxidation process to study the changes in their growth rate. Two species of algae *Nostoc muscorum* and *Spirulina* sp. were studied in a petri dish of diameter 100 mm. DBD plasma is produced between the 90 mm diameter copper electrodes and petri dish placed between the electrodes. The high voltage power supply produced uniform DBD plasma at 15 kV, 20 kHz between the electrodes and the algae samples were exposed between electrodes for 300 s (Fig. 2).

Due to the presence of chlorophyll, the algae culture shows maximum absorbance at 684 nm. Therefore a correlation between cell count (obtained by using haemocytometer) and optical density at 684 nm (OD_{684}) of the cell suspension for each species was determined. The OD_{684} of two species of algae *Nostoc muscorum* and *Spirulina* sp. were measured by a UV-vis spectrophotometer (Spectroquant® UV/VIS

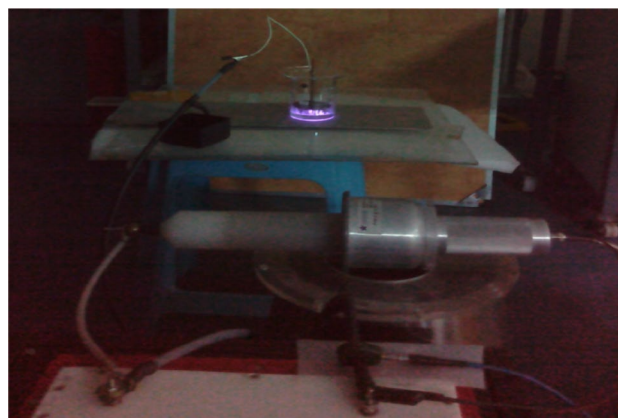


Fig. 2 DBD plasma exposure to algal species

Table 2 Optical density measured and cell reduction of algal species

Day	<i>Algae species: Nostoc muscorum</i>			<i>Algae species: Spirulina sp.</i>		
	Control	Optical density	Cell reduction (%)	Control	Optical density	Cell reduction (%)
1	0.275	0.153	44.36	0.626	0.428	31.63
2	0.316	0.139	49.45	0.646	0.384	38.66
3	0.351	0.123	55.27	0.669	0.698	48.1
4	0.335	0.092	66.54	0.665	0.547	58.94
5	0.318	0.064	76.73	0.667	0.523	66.93

Spectrophotometer Pharo 300-Merck) using Milli-Q water as the blank. Therefore, the OD₆₈₄ was used as a measure to monitor the changes in cell concentration and cell growth (Table 2).

The cell number change (%) can be calculated by using Eq. 3:

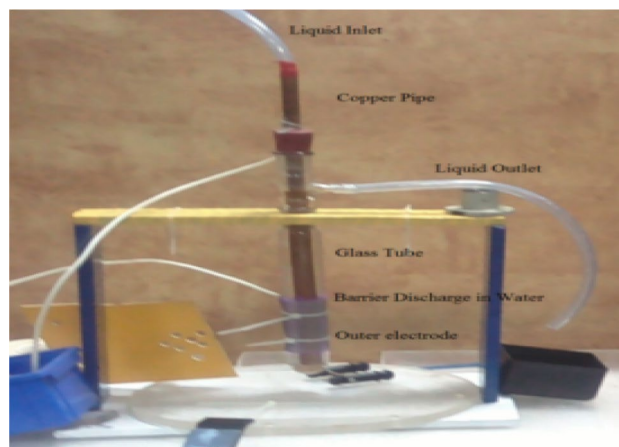
$$\text{Cell reduction(\%)} = \frac{\text{OD}_{684} \text{ before treatment} - \text{OD}_{684} \text{ after treatment}}{\text{OD}_{684} \text{ before treatment}} \times 100\% \quad (3)$$

It can be seen was that DBD plasma is very much effective for cell reduction and growth inhibition for algal mass. The UV, ozone and other reactive species produced during the plasma discharges played the major role in damaging the cell structure and hinder their growth.

Breakdown of Complex Phenol

Degradation studies on complex phenol mixed with water were carried out using pulsed DBD produced by high voltage power supply. The treatment setup has 10 mm diameter copper pipe placed inside the 20 mm glass test-tube (Fig. 3). Copper tube will also act as a ground electrode and the high voltage terminal is connected at the outer electrode made up of SS mesh. The water flow inside the copper tube and then comes out at the top of the tube. High voltage pulses of 15 kV, 20 kHz are applied across the electrodes, which produces intense dielectric barrier discharge over the surface.

The phenol mixed water is passed through the DBD plasma and then collected. Samples of three different initial concentration of phenol 10 mg/l, 50 mg/l and 100 mg/l, were used and treated for 1 min and 5 min (Table 3). The concentration of phenol was measured using UV-spectrophotometer.

**Fig. 3** DBD plasma treatment setup for a breakdown of complex phenol**Table 3** Phenol concentration before and after treatment

Phenol concentration (mg/l)	Phenol concentration (mg/l) after treatment	
	Time = 1 min	Time = 5 min
10	9	7
50	46	32
100	91	62

$$\% \text{Degraradation of Phenol} = \frac{C_0 - C_f}{C_0} \times 100 \quad (4)$$

It can be seen that after passing the phenol through the DBD plasma, its concentration is reduced. The reduction in the concentration of phenol is due to the breakdown of phenols by UV, ozone and reactive species produced due to DBD plasma. It was also seen that by increasing the exposure time will further break the phenol in water.



Fig. 4 DBD quartz tube with power supply



Fig. 5 Intense UV generation from the quartz tube

UV, Ozone and Reactive Species Generation with DBD Plasma discharges in Quartz Tube

The 45 kV, 50 kHz high voltage power supply is connected to the 0.5 mm copper foil and copper mesh electrodes and evacuated quartz tube is used as a dielectric barrier

between the electrodes. The inner and outer diameters of the quartz tube are 50 mm and 70 mm. The thickness and length of the quartz tube are 1 mm and 300 mm (Fig. 4). The voltage and frequency is slowly increased and the DBD plasma is generated after 8 kV and 10 kHz.

For the present evacuated quartz tube the maximum surface area of DBD plasma is seen at 12 kV, 30 kHz power supply rating. This DBD plasma generates intense UV radiations, ozone and other reactive species in ambient air (Fig. 5).

DBD Sterilization System

A portable DBD plasma-based sterilization system is developed for fighting the Covid-19 pandemic. It can sterilize and disinfect the microorganism contaminated surfaces with UV, ozone and short-lived molecules of metastable states and excited chemical species produced during the DBD plasma discharges in ambient air. The DBD sterilization system consists of a pressurized chamber and dielectric quartz tube of 1 mm thickness, 25 mm ID and 300 mm length and copper-foil mesh electrodes connected to pulse power supply (Fig. 6). The sterilization chamber can be pressurized with ambient air up to 2 atm. A SS mesh is placed above the DBD tube to keep the contaminated substances on it for disinfection/sterilization.

When the power supply is switched on and high voltage, high frequency pulses of 17 kV, 30 kHz are applied across the tube; it will produce non-thermal plasma of partially ionized gas using DBD (Fig. 7). This plasma generates energetic ions and electrons which can reduce the concentrations of microorganism and also destroy pathogens such as bacteria, virus, fungi and prions in the sterilization system. A small fraction of air generates this plasma, the rest of the molecules are at ambient temperature. This plasma produces very energetic ions and electrons, which can produce large quantities of short-lived molecules of metastable and excited states and chemical species such as H, N and O atomic species, OH and ON radicals, ozone, nitrous and nitric acids. The optical spectrometry of DBD air plasma confirms the 312 nm, 376 nm, 423 nm optical emissions of excited Oxygen and 333 nm, 353 nm, 395 nm optical emissions of Nitrogen species (Fig. 8). The sterilization of microorganisms on the contaminated surface is achieved by bombarding them with UV, electrons, ions and short-lived chemical species and also by the oxidizing effect of the highly reactive chemical species in the plasma.

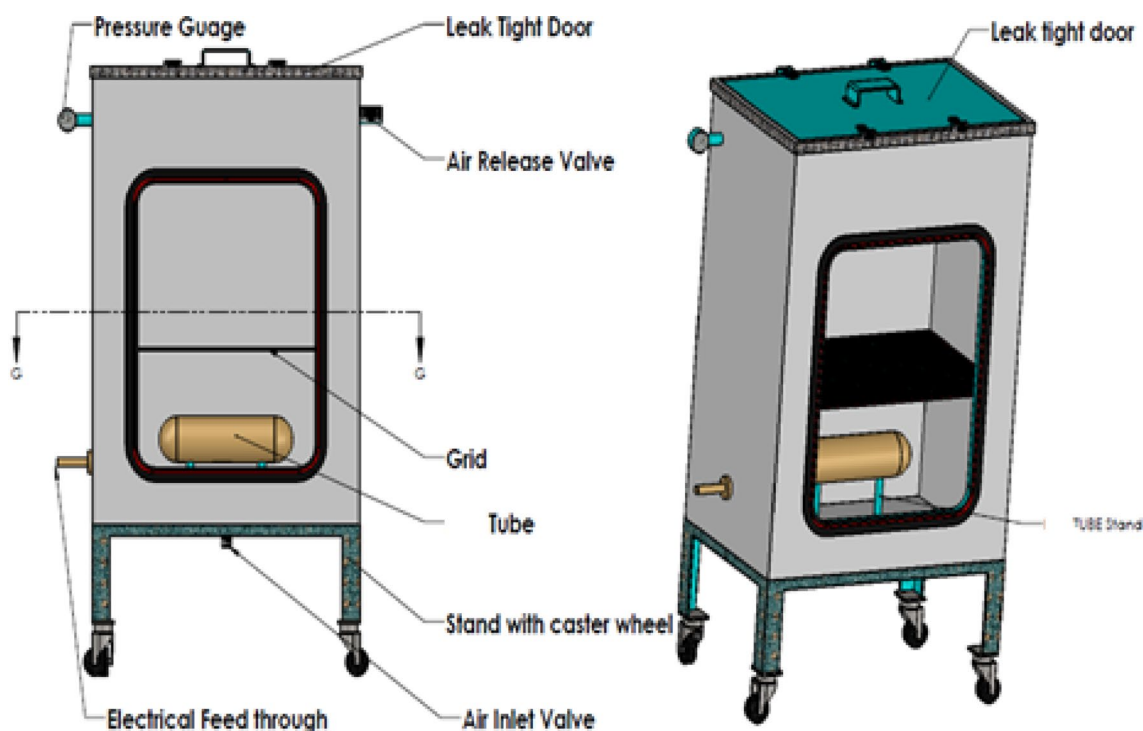


Fig. 6 DBD plasma-based sterilization system



Fig. 7 DBD plasma with ambient air

Conclusion

A 45 kV, 50 kHz power supply was developed and tested to produce DBD plasma in air. This non-thermal plasma produces intense UV, ozone and short-lived molecules of metastable and excited states species of nitrogen and oxygen in ambient air, which can be used for growth control in algae, treatment of chemical wastewater. A portable

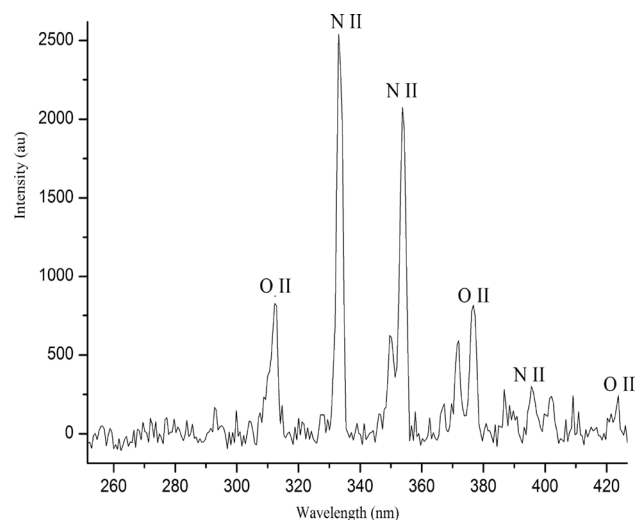


Fig. 8 Optical spectrometry of DBD air plasma

sterilization system is made with DBD plasma produced with 1 mm thick quartz tube and foil-mesh electrodes operating at 17 kV, 30 kHz for sterilization and treatment of contaminated surfaces, garments and used disposable protective gears to break the chain of viral transmission for fighting Covid-19 pandemic.

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References

- Gallagher M, Vaze N, Gangoli S, Vasilets VN, Gutsol AF et al (2007) Rapid inactivation of airborne bacteria using atmospheric pressure dielectric barrier grating discharge. *Plasma Sci IEEE Trans* 35(5):1501–1510
- Kim Y, Sewhan J, Gook-Hee H, Gi CK, Jin JC, Eun HC, Han SU, Guangsop C (2015) Plasma apparatus for biomedical applications. *IEEE Trans Plasma Sci* 43(4):944–950
- Kogelschatz U, Eliasson B, Egli W (1997) Dielectric-barrier discharges. Principle and applications. *J Phys IV Colloque* 7:47–66
- Laroussi M (2005) Low temperature plasma-based sterilization: overview and state-of-the-art. *Plasma Process Polym* 2(5):391–400
- Mastanaiah N, Johnson JA, Roy S (2013) Effect of dielectric and liquid on plasma sterilization using dielectric barrier discharge plasma. *PLoS ONE* 8(8):e70840. <https://doi.org/10.1371/journal.pone.0070840>
- Moisan M, Barbeau J, Moreau S, Pelletier J, Tabrizian M et al (2001) Low-temperature sterilization using gas plasmas: a review of the experiments and an analysis of the inactivation mechanism. *Int J Pharm* 226(1):1–21
- Surender KS, Anurag S (2015) Development of compact rapid charging power supply for capacitive energy storage in pulsed power drivers. *Rev Sci Instrum* 86(2):023503
- Surender KS, Anurag S (2016) Design and testing of 45 kV, 50 kHz pulse power supply for dielectric barrier discharges. *Rev Sci Instrum* 87:105115
- Tanion M, Wang X, Takashima K, Katsura S, Mizuno A (2005) Sterilization using dielectric barrier discharge at atmospheric pressure. In: *Fortieth IAS Annual Meeting. Conference Record of the 2005 Industry Applications Conference*, 2005. Doi: 10.1109/IAS.2005.1518421
- Xia T, Kleinheksel A, Lee M, Qiao Z, Wigginton KR, Clack HL (2019) Inactivation of airborne viruses using a packed bed non-thermal plasma reactor. *J Phys D Appl Phys* 52:255201

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