This is a peer-reviewed, accepted author manuscript of the following item:
Huang, G., Wang, T., Timoshkin, I., MacGregor, S., Given, M., & Wilson, M. (2012).
Investigation of ozone generation using dielectric barrier discharges at 50 Hz, 2.6 kHz and 20 kHz. In
Proceedings of the XIX International Conference on Gas Discharges and Their Applications (pp. 650-653).
High Voltage Engineering

INVESTIGATION OF OZONE GENERATION USING DIELECTRIC BARRIER DISCHARGES AT 50 Hz, 2.6 kHz AND 20 kHz

G.M. Huang, T. Wang, I.V. Timoshkin, S.J. MacGregor, M.J. Given, M.J. Wilson

Department of Electronic and Electrical Engineering, University of Strathclyde, Royal College Building, 204 George street, Glasgow, UK, G1 1XW.
E-mail: tao.wang@strath.ac.uk

ABSTRACT

Experiments were conducted to investigate ozone generation (DBD) at different frequencies. A cylindrical DBD ozone generator with a discharge gap of 0.3 mm has been developed. 50 Hz AC power was used to provide power density of 44.2 W/m^2. 2.6 kHz and 20 kHz AC energisation frequencies were employed to provide power densities of 2.37 kW/m^2 and 19.08 kW/m^2 respectively. Discharge current, optical emission signals and discharge power were obtained under three frequencies. Ozone concentration and production efficiency at different feed gas flow rates were measured and calculated. It was found that discharge mode was different for positive and negative half-cycle of the applied voltage. Results show that ozone production efficiency increases with an increase in the ozone concentration at 50 Hz, however this efficiency decreases with an increase in the ozone concentration at 2.6 kHz and 20 kHz. For the same ozone concentration level, 2.6 kHz is more efficient than 20 kHz. The highest ozone production efficiency achieved in this work is 191.5 g/kWh at 50 Hz and the highest ozone concentration is 271 g/Nm^3 at 2.6 kHz.

1. INTRODUCTION

Ozone is a strong oxidation agent. It is currently being used in the wide range of applications including water treatment, organic degradation and pulp bleaching [1]. Ozone has higher oxidation-reduction potential (2.07 V) as compared with other oxidants including chlorine (Cl, 1.36 V) and hydrogen peroxide (H_2O_2, 1.78 V). Ozone is also environmental friendly agent which produces minimum chemical by-products.

Current industrial ozone applications require advanced ozone generators that can provide high concentrations of ozone at high production efficiencies. Power density, ozone concentration and ozone production efficiency are the main parameters used for evaluation of the performance of industrial ozone generators. However, these parameters may result in conflicting requirements [2]. It is the interest of this work to investigate the relationship between ozone production efficiency and ozone concentration under different power density levels using three different energisation frequencies, 50 Hz, 2.6 kHz and 20 kHz. Discharges parameters at these frequencies were measured and discussed.

2. EXPERIMENT SET-UP

The dielectric barrier discharge (DBD) ozone generator with cylindrical electrodes configuration has been developed. Its schematic diagram is shown in Fig.1. Three borosilicate glass tubes of 300 mm in length were used as dielectric barriers. The tubes have an outer diameter of 15 mm and thickness of 1.2 mm. The active discharge length of the reactor is 240 mm. These glass tubes are placed inside a stainless steel pipe with water surrounding as grounding electrodes. Stainless steel rods served as high voltage electrodes with diameter of 12 mm were located inside each glass tube. The discharge gap between the dielectric barrier and high voltage electrode was 0.3 mm. The temperature of circulating water was kept at a constant value of $15^{o}\mathrm{C}$ in the experiment.

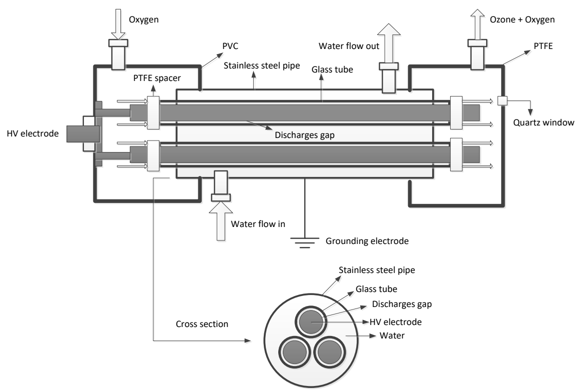


Fig. 1 Schematic diagram of ozone reactor

The experimental system is shown in Fig. 2. The gas used is 99.99% oxygen from BOC with a dew point $-40^{o}\mathrm{C}$. Gas flow rate is measured by ALICAT M series mass flow meter. Ozone concentration is monitored by IN-USA Mini-HiCon Ozone Analyzer. A regulating transformer followed by a step up high voltage transformer is used as the 50 Hz power supply. The output voltage has a peak value of 6 kV. A thyristor controlled frequency inverter is employed to provide high frequency AC power. A sinusoid voltage with a peak value of 6 kV and frequencies of 2.6 kHz and 20 kHz were generated using the frequency control of the inverter. The output power of the inverter was adjusted by power density modulation (PDM) control.

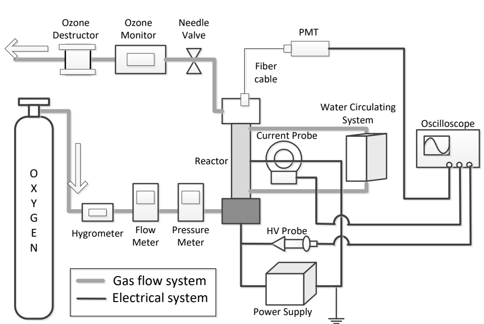


Fig. 2 Schematic diagram of the experimental system

The applied voltages were measured by Tektronix P6015A high voltage probe (bandwidth 75 MHz , division ratio 1000:1) and the discharge current was measured by Pearson current probe (Model 6585) with the bandwidth from 400 Hz to 250 MHz. A photomultiplier (ET 9125B) was employed to detect emitted light from discharges. A quartz window is placed parallel to the cross section of discharges space.

A Tektronix digital oscilloscope (TDS 3054 B) with the bandwidth of 500 MHz and sample rate of 5GS/S was used to display and record the voltage, current and PMT signals.

All the experiments in this work were carried out at atmospheric pressure.

3. RESULTS AND DISCUSSION

3.1. DISCHARGE CURRENT AND PMT SIGNAL MEASUREMENT.

Discharge current and photomultiplier signals were measured and their results are shown below in Fig. 3.

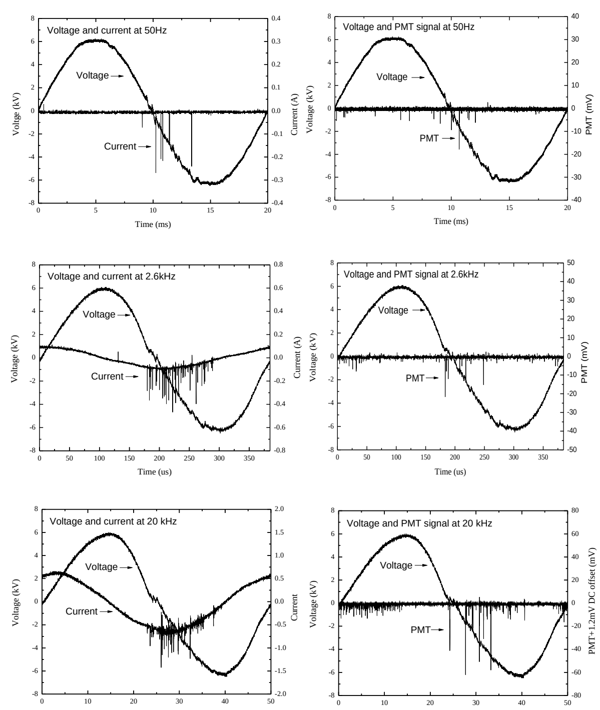


Fig. 3 Discharge current and PMT signal at different frequencies

Results show that the current across the ozone reactor contains two components: sinusoidal continuous current and transient current. Every PMT peak signal corresponds to one transient impulse event (impulsive current) which results from the plasma filaments developing near the position where PMT is placed. As shown from the above figure, higher frequencies produce larger transient current amplitudes and larger PMT signals on average. This may result from an increased number of micro discharges at higher frequency energisation. It can be observed that for all tested frequencies the transient currents are significantly lower during the positive voltage half-cycle as compared with that during the negative voltage half-cycle. From the optical measurements, it can be seen that PMT signals during the positive half-cycle have low amplitude but distributed more homogeneously as compared with larger magnitude signals during the negative half-cycle. The reason for this may result from the pulsed glow-like discharge mode during the positive half-cycle and filamentary discharge mode during the negative half-cycle. When the polarity of applied voltage is positive, there are a large number of pre-ionization electrons deposited uniformly on the surface of dielectric barrier from the previous half-cycle. Large electron density causes the overlap and coalescence of primary avalanches resulting in a pulsed glow-like discharge [3, 4]. During the positive half-cycle, the electrons are neutralized by positive high voltage electrode so when the polarity of the high voltage electrode reverses to negative, the density of electrons left in the discharge space is much lower. So the discharge shows filamentary mode during this negative half-cycle. Currently, the effects of polarity of voltage stress on the ozone generation process are not fully understood and required further investigation.

3.2. DISCHARGE POWER MEASUREMENT

Discharge power was obtained by a measuring capacitor of 235nF connected in series with the ozone reactor. The power was calculated by the following equation:

$$P = \frac{1}{T}\int_0^T vidt = \frac{1}{T}\int_0^T v\frac{dq}{dt}dt = \frac{1}{T}\int_0^T vdq \quad (1)$$

Below is the typical Q-V trace figure achieved from the experiment:

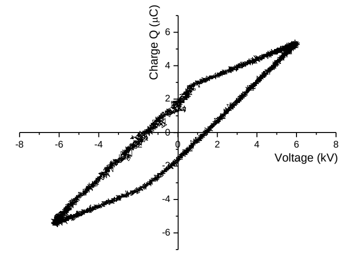


Fig. 4 Lissajous figure

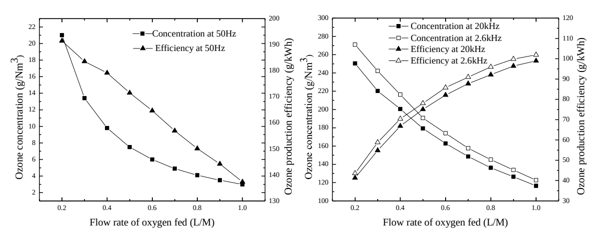
In this work, 12% PDM was applied to the 20 kHz energisation to achieve approximately same average power as at 2.6 kHz. The average power density values under different energisation frequencies were calculated by dividing the average discharge power by the discharge area and results of this analysis are shown in table 1 below:

Table 1. Average power density at different frequencies

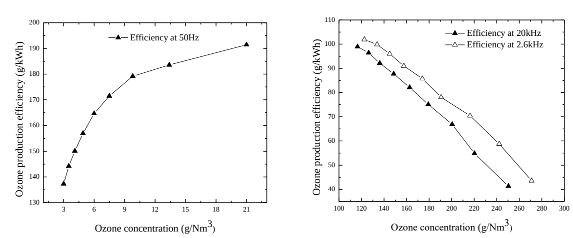
Power supply	50 Hz	2.6 kHz	20 kHz
Average PD	44.2 W/m^2	2.37 kW/m^2	2.29 kW/m^2

3.3. OZONE PRODUCTION

Ozone concentration and production efficiency under three frequencies are presented in Fig. 5.



(a)



(b)

Fig. 5 (a) Ozone concentration and production efficiency versus different flow rate (b) Ozone production efficiency versus different ozone concentration

Fig. 5(a) shows that ozone concentration decreases with an increase of feed gas flow rate for all tested frequencies. The highest ozone concentrations achieved at 50 Hz, 2.6 kHz and 20 kHz are 21 g/Nm^3, 271 g/Nm^{3}and 250 g/Nm^3 respectively. As shown in Fig. 5(a), the ozone production efficiency drops with the increase in the feed gas flow rate at 50 Hz energisation, while the efficiency rises with the feed gas flow rate at high frequencies of 2.6 kHz and 20 kHz. The highest ozone production efficiency achieved at 50 Hz is 191.5 g/kWh at the flow rate of 0.2 L/M while the highest efficiency achieved at 2.6 kHz and 20 kHz are 101.9 g/kWh and 99.0 g/kWh respectively at the flow rate of 1 L/M. The lower efficiency in the kHz excitation range should be related to corresponding high ozone concentrations, because the dissociative attachment of electrons in ozone increases with an increase in the concentration of ozone [5, 6]. As shown in Fig. 5 (b), the ozone production efficiency increases with the increase in the ozone concentration at 50 Hz while at 2.6 kHz and 20 kHz energisation frequencies, the ozone production efficiency decreases with an increase in the ozone concentration. The reason for this may be due to the fact that the ozone dissociation rate is negligible at low background ozone concentration but it becomes comparable with ozone generation rate when the background ozone concentration rises to a high level. From Fig 5(b), it can be seen that at the same ozone concentration, energisation frequency of 2.6 kHz produces slightly higher ozone production efficiency as compared with the energisation frequency of 20 kHz. At the typical ozone concentration 150 g/Nm^3 used in industrial applications, the efficiency at 2.6 kHz is 94.2 g/kWh while at 20 kHz the efficiency is 87.2 g/kWh.

4. CONCLUSION

The electrical parameters of dielectric barrier discharges used for ozone generation under three energisation frequencies of 50 Hz, 2.6 kHz and 20 kHz were measured and analyzed in this work. For all tested frequencies different discharge modes were observed for negative and positive half-cycles of the voltage stress. The reason for this probably relates to the electrode structure of DBD ozone generator that has only one electrode covered by the dielectric material. Results of ozone production show that at low power density provided by 50 Hz energisation frequency ozone production efficiency increases with the ozone concentration. But at high power densities provided by kHz excitation ranges, the ozone production efficiency drops with the ozone concentration. Results also show that 2.6 kHz is slightly more efficient than 20 kHz.

REFERENCE

[1] R.G.Rice and A.Netzer, "Handbook of Ozone Technology and Applications," vol. 1, 1982.

[2] U. Kogelschatz, B. Eliasson, and M. Hirth, "Ozone generation from air and oxygen," *OZONE SCIENCE & ENGINEERING*, vol. 10, pp. 367-378, 1988.

[3] U. Kogelschatz, "Filamentary, patterned, and diffuse barrier discharges," *IEEE Transactions on Plasma Science,* vol. 30, pp. 1400-1408, 2002.

[4] R. Jozef and M. S. Daniel, "The transition from a filamentary dielectric barrier discharge to a diffuse barrier discharge in air at atmospheric pressure," *Journal of Physics D: Applied Physics,* vol. 38, pp. 547-554, 2005.

[5] P. Tegeder, P. A. Kendall, M. Penno, N. J. Mason, and E. Illenberger, "Electron stimulated desorption of O– and O2– from condensed ozone: possible implications for the heterogeneous photochemistry of stratospheric O3," *Physical Chemistry Chemical Physics,* vol. 3, pp. 2625-2629, 2001.

[6] C. W. Isobel, M. G. John, J. M. Nigel, and M. George, "Dissociative electron attachment (DEA) in ozone 0 - 10 eV," *Journal of Physics B: Atomic, Molecular and Optical Physics,* vol. 29, pp. 4749-4759, 1996.