Optimization of DBD Reactors for Ozone Generation

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Abstract Ozone generation efficiency is often used as an indicator to evaluate the performance of DBD reactors. This study aims to provide a general guideline for designing cylindrical DBD reactors with better performance based on the ozone generation efficiency. In addition to experiments, a numerical model is developed in this study. Specific energy density, defined as the ratio of deposited power to flow rate, is an important parameter for plasma. Two breakdown mechanisms are adopted to predict the breakdown voltage of a specific reactor to determine the theoretical deposited power. The results of experiments and simulation show good consistency, verifying the validity of the modeling. Based on the experimental and simulation results, smaller reactor and larger inner electrode will achieve better energy efficiency at the same specific energy density in that the mean electron energy is higher so that most of the energy can be used to produce atomic oxygen, the precursor for ozone formation. However, for reactors with small diameters, there exists an optimum inner electrode diameter. The influence of dielectric thickness is also investigated. Increasing the dielectric thickness of a specific reactor results in higher breakdown voltage, leading to lower deposited power and ozone concentration at a fixed applied voltage.

Keywords : dielectric barrier discharge, ozone generation, reactor design

1. INTRODUCTION

Dielectric barrier discharge (DBD), initially developed for ozone generation, has been under intensive investigations for removing a variety of gaseous pollutants, such as VOCs[1-2], NOx[3], SOx[4], during the last two decades. More recently, modifications of conventional DBD reactors including packed-bed reactors and combined plasma catalysis are proposed and have been successfully demonstrated to achieve better performance[5-8]. However, the pressure drop and difficulty to release heat caused by packing dielectric pellets or catalysts inside DBD reactors are the main disadvantages.

Various parameters have influence on DBD reactor performance. In general, these parameters can be classified into three categories including reactor, electrical and gas parameters. Knowing how each parameter affects the DBD reactor performance would be of much use. However, the complexity of plasma characteristics makes it a tough job to optimize the performance without assistance of numerical modeling. Ozone generation efficiency is often used as an indicator to evaluate the performance of DBD reactors. The purpose of this study is to build up a numerical modeling that can assist the design of DBD reactors. The influence of reactor geometry (diameter of inner and outer electrode and thickness of dielectric) on ozone production are investigated in this study. Experimental and simulation results show great consistency, confirming the validity of the modeling. The results obtained by this modeling provide useful information for designing a DBD reactor for ozone generation. In the future, the same modeling will be used for other applications like removing gaseous pollutants, hoping to offer a general guideline for DBD reactor design.

2. EXPERIMENTAL

Nine different cylindrical DBD reactors were used to investigate the influence of reactor geometry on ozone generation performance. The thickness and material of dielectric for all DBD reactors investigated were 2 mm and pyrex glass, respectively. The radii of inner electrode included 0.1, 0.2 and 0.3 cm. Glass tubes with radius of 0.6, 1.1 and 1.6 cm were adopted. Oxygen used for ozone generation was supplied from a gas cylinder having a purity of 99.9%. The ozone concentration was measured with an ozone monitor (Sorbios, Model OMH, 100.2/EG-200).

The high-voltage power supply was composed of a power meter (Chen Hwa Co., Model 2100) and a high-voltage transformer (Jui-Hsiang PTY Co. Ltd.). The applied voltage and frequency were controlled by the power meter. For all experiments conducted in this study, the applied frequency was kept at 100 Hz. As for the deposited power, the Tektronix
3014B digital oscilloscope equipped with a high voltage probe (Tektronix P6015A) and a voltage probe (Tektronix 613B) was employed to measure the Lissajous Figure (V-Q plot). A 1-μF capacitor was connected in series with DBD reactor to measure the charge.

Generally, the ozone concentration required an amount of time to reach steady, depending on the operation conditions. All the data represented in this study were the steady-state values.

3. MODELING DESCRIPTION

Lissajous figure is widely used to measure the deposited power of DBD reactors. The area enclosed by the parallelogram stands for the energy consumed per cycle of alternative current. Consequently, the deposited power can be described by the following equation:

\[
\gamma \left( \int (\alpha - \eta) \, dr \right) - 1 = 1 \quad [4]
\]

where \( \gamma \) is the secondary emission coefficient for the electrode, \( \alpha \) is Townsend first ionization coefficient (cm\(^{-1}\)), \( \eta \) is the electron attachment coefficient (cm\(^{-1}\)), and \( r \) is the radial distance (cm). However, at higher \( pd \), the breakdown mechanism turns into streamer breakdown. For this breakdown mechanism, the criterion for producing plasma is that the space field at a certain position in the gap must be higher than its corresponding external field, which can be expressed as\(^{[11]}\):

\[
\frac{e_0 \exp \left( \int \alpha_{eff} (r) \, dr \right)}{4\pi \cdot \varepsilon_s} > E(\xi) \quad [5]
\]

where \( \varepsilon_0 \) is the charge of an electron (1.6x10\(^{-19}\) C), \( \varepsilon_s \) is the relative permittivity of background gas, \( \alpha_{eff} \) is the effective ionization coefficient (cm\(^{-1}\)), namely, the difference between \( \alpha \) and \( \eta \), \( D_s \) is the diffusion coefficient of electrons (cm\(^2\) s\(^{-1}\)), \( v_{drift} \) is the drift velocity of electrons (cm s\(^{-1}\)), and \( E(\xi) \) is the applied field at the position apart from the outer surface of inner electrode with a distance of \( \xi \). For a cylindrical DBD reactor, the electrical field is not uniformly distributed in the reactor can be expressed as:

\[
\frac{\varepsilon_s}{\varepsilon_d} \left( \ln \frac{b}{r_i} \right) = \frac{2 \pi \varepsilon_d l_i}{\ln \left( \frac{b}{r_i} \right)} \quad [1]
\]

where \( C_d \) is the capacitance of gas gap (F), \( C_g \) is the capacitance of gas gap (F), \( \tilde{V} \) is the peak value of applied voltage (V), and \( V_{min} \) is the minimum external voltage to initialize discharge (V). This is also known as Manley equation\(^{[9]}\). The theoretical capacitances of gas gap \( C_g \) and dielectric layer \( C_d \) for a cylindrical DBD reactor can be expressed as:

\[
C_g = \frac{\varepsilon_s \times l}{\rho \cdot \ln \left( \frac{b}{r_i} \right)} = \frac{2 \pi \varepsilon_d l}{\ln \left( \frac{b}{r_i} \right)} \quad [2]
\]

\[
C_d = \frac{\varepsilon_s \times l}{\rho \cdot \ln \left( \frac{b}{r_o} \right)} = \frac{2 \pi \varepsilon_d l}{\ln \left( \frac{b}{r_o} \right)} \quad [3]
\]

where \( l \) is the discharge length (cm) and \( \varepsilon_s \) and \( \varepsilon_d \) are the relative permittivities of background gas and dielectric, respectively. The meaning of \( r_i, b \) and \( r_o \) are illustrated in Fig. 1.
The electrical field in the gap induced by external voltage can be determined by the following equation:

$$E_g = \frac{V_g}{r \ln \left( \frac{b}{r_i} \right)}$$ \[6\]

where \(V_g\) is the voltage across the gap. Therefore, the parameters including \(\alpha, \eta, D,\) and \(v_{\text{drift}}\) are all functions of radial distance. All these parameters and electron energy distribution function (EEDF) are calculated with BOLSIG code\[12\]. In this study, both breakdown mechanisms were adopted to simulate to examine which one is more suitable for a specific reactor.

Once the plasma is produced in the DBD reactor, the voltage across the gap remains constant\[13\]. On the other hand, the charges transferred by an individual microdischarge is constant regardless of applied voltage\[14\]. It implies that the change of applied voltage only affects the number of microdischarges rather than the charge transferred by each microdischarge. Based on the above-mentioned phenomena, it is assumed that the mean electron energy is not a function of applied voltage for a specific reactor and gas composition. In this study, the mean electron energy is determined as the following equation:

$$\bar{T}_e = \int_T T_e(r) 2\pi dr. \quad [7]$$

The concept of PFR (plug flow reactor) is adopted to develop a one-dimensional modeling in this study. Assumptions made are described as follows. Taking the statistical distribution of microdischarges into consideration, it is reasonable to treat the DBD reactor during discharge as a serial combination of microdischarge region and nondischarge region which alternate with each other along the axial direction of the reactor. The electron energy is assumed to be constant throughout the microdischarge and the deposited power is equally allotted to each microdischarge. The radial variations in concentrations of species are not taken into account. Besides, the reactor is assumed to be isothermal, namely temperature variation with time and position is not considered.

The reaction set adopted in this study includes 65 reactions (26 electron-impact, 26 ion-involved, 13 neutral-neutral reactions). All the electron-impact reactions are expressed as function of electron temperature to reflect the influence of reactor size on plasma characteristics. The total species amounted to 10. The loss of elementary balance for each run is rather low, typically smaller than \(10^{-6}\), which indicates that no truncating error occurs during computing process. Hence, reliability of the predicted results is certified.

4. RESULTS AND DISCUSSION

The purpose of this study is to provide a general guideline for DBD reactor design based on the energy efficiency for ozone generation. In addition to experiments, a numerical modeling is developed to simulate the plasma characteristics in reactors with different sizes.

The influences of \(b\) and \(r_i\) on the energy efficiency for ozone generation are shown in Fig. 2 and Fig. 3, respectively. All the data shown in Fig. 2 and Fig. 3 were obtained from experiments. When \(r_i\) is fixed, the energy efficiency increases with decreasing \(b\) at the same specific energy density. The trend is monotonous for all the reactors tested in this study. On the other hand, larger \(r_i\) results in higher energy efficiency while \(b\) is fixed, as shown in Fig. 3 (a) and (b). However, there exists an optimum \(r_i\) when \(b\) is as low as 0.4 cm (see Fig. 3 (c)). One possibility causing this result is that the temperature of the reactor with \(r_i = 0.3\) cm is higher than those of the other two reactors shown in Fig. 3 (c) at the same specific energy density, accelerating decomposition rate of ozone.

![Fig. 2. Influence of \(b\) on the energy efficiency of ozone](image-url)
generation (a) $r_i = 0.1$ cm, (b) $r_i = 0.2$ cm and (c) $r_i = 0.3$ cm.

Specific power density, namely the ratio of deposited power to flow rate, is an important parameter for plasma. The theoretical deposited power can be determined through equation [1]. For a reactor with given size, the only unknown in equation [1] is the breakdown voltage $V_{\text{min}}$. Two breakdown mechanisms are adopted to predict the breakdown voltage for a specific reactor. Fig 4. shows the results obtained based on Townsend and streamer breakdown mechanisms. When the value of $pd$ is lower than 200 Torr·cm, the breakdown follows Townsend criteria\cite{15}. In this study, all the experiments were conducted at atmospheric pressure. In this case, the gap should be smaller than 0.26 cm to keep the product of $p$ and $d$ lower than 200 Torr·cm. As can be seen in Fig. 4, the simulated breakdown voltages based on streamer breakdown mechanism show better precisions than those based on Townsend breakdown.

The mean electron temperature of various reactors simulated based on two breakdown mechanisms are shown in Fig. 5. The net electrical field for streamer breakdown is assumed to be the sum of external electrical field and space field. Hence, the electron temperature obtained from streamer breakdown is higher than that from Townsend breakdown. Obviously, smaller $b$ and larger $r_i$ lead to higher mean electron temperature.

Fig 6. displays the energy consumption partition at mean electron temperature ranging from 1 to 5 eV of several electron-impact reactions including:
\begin{align*}
e^+O_2 &\rightarrow O^+ + O \quad (1) \\
e^+O_2 &\rightarrow e^+ + 2O \quad (2)
\end{align*}
Here, \( \text{O}_2^* \) represents all the excited species of oxygen molecules. The energy consumed by reaction (6) shows significance at lower electron temperature. As the electron temperature increases, more energy is consumed by reaction (2) and (7). Since reaction (2) and (7) can produce \( \text{O} \) radicals, the precursor for ozone formation, it can be expected that the ozone generation efficiency will be higher when the electron temperature increased. As mentioned above, smaller \( b \) and larger \( r_i \) results in higher mean electron temperature. It implies that the reactor with smaller \( b \) and larger \( r_i \) will achieve higher energy efficiency. This is consistent with the experimental data, as shown in Fig. 2 and 3.

**Fig. 6.** Energy consumption partition for several electron-impact reactions as a function of mean electron temperature.

**Fig. 7** (a) and (b) show the simulation results for the reactors with \( b = 0.9 \) cm and \( b = 0.4 \) cm, respectively. Fig. 7 (a) is based on streamer breakdown mechanism since all the values of \( pd \) for the reactors are greater than 200 Torr cm. On the contrary, Fig. 7 (b) is based on Townsend breakdown. All the simulation results show good consistency with the experimental data, confirming the validity of the numerical modeling developed in this study.

**Fig. 8** shows the influence of dielectric thickness on the deposited power and ozone concentration obtained from simulation. For a cylindrical DBD reactor, the electrical field in the gap is not a function of \( r_o \) based on equation [6]. It implies that the voltage across the gap required to initiate plasma stays the same when \( r_o \) is changed. However, the dielectric thickness affects the ratio of \( V_g \) to \( V_d \) (voltage across the dielectric). Based on equation [6], increasing \( r_o \) results in a lower \( C_d \); therefore, the breakdown voltage will be higher when the dielectric thickness is increased. That’s reason why the deposited power and ozone concentration decreases with increasing dielectric thickness at the same applied voltage. This result is consistent with the experimental data of Garamoon et al.\[^{[16]}\], verifying the validity of the numerical modeling developed in this study.
Fig. 8. Influence of dielectric thickness on (a) deposited power and (b) ozone concentration.

5. CONCLUSION

A general guideline for DBD reactor design is provided based on both experiments and simulation. The results obtained from experiments and simulation show good consistency. Reactors with smaller reactor diameter and larger inner electrode result in higher electron temperature and better energy efficiency. However, when the reactor is too small, there exists an optimum inner electrode diameter. One possibility is the higher temperature accelerates the ozone decomposition rate. Increasing the dielectric thickness of a specific reactor results in higher breakdown voltage, leading to lower deposited power and ozone concentration at fixed applied voltage.

References