Electrical and Chemical Characteristics of Dielectric Barrier Discharge Process for Environmental Applications

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Abstract

There are intensive studies on applications of dielectric barrier discharges (DBD) for the treatment of flue gas emitted from various pollution sources. In this experiment, electrical, physical, and chemical parameters were varied systematically in an attempt to understand how the reactor design and electric parameters affect the energy transfer and pollutant removal characteristics in the DBD process. Experimental parameters were the radius of discharge electrode, initial concentrations, surface roughness of discharge electrode, and others. The average dissipation energy increased as the peak voltage increases. The sparking potential increased linearly with the larger gap distance, and the sparking potential for the coarse electrode was lower than that for the smooth electrode. The optimum gap distances for getting the highest dissipation energy in a given voltage were 7 cm and 9 cm at low and high voltages, respectively. NO was converted to NO\textsubscript{2} rapidly at a low energy density. The energy density needed to obtain the same conversion rate increased as the initial concentration increases. It was observed that the NO conversion was insensitive to the residence time. The NO conversion rate for the smooth electrode was higher than that for the coarse electrode at the same energy density. The NO conversion rate of the discharge electrode with 35 mm radius was higher than that of the discharge electrode with 30 mm radius at the same residence time and the same energy density. The intensity of light emission increased as the increase of the dissipation energy (or peak voltage) and the flow rate.

1. Introduction

Nitrogen oxides are major air pollutants, and they are produced in all combustion processes. The major part of the emissions comes from motor vehicle exhausts, power plants, and industry. As nitrogen oxides and their related products are highly toxic to human and hazardous to the environment, their emission should be reduced as much as possible, and the development of the NOx control technologies becomes an important issue. There are intensive investigations on the application of non-thermal plasmas combined with a catalytic system for the treatment of NOx emitted from various pollution sources. Non-thermal plasma gives an efficient mean for selective partial oxidation of NO to NO\textsubscript{2}. Pre-converting NO to NO\textsubscript{2} opens the opportunity for a wider range of the selective catalytic reduction catalysts and improves the durability of these catalysts.\textsuperscript{(1)} The dielectric barrier discharge (DBD) is one of non-thermal plasma techniques and offers the advantage to excite molecules to reaction processes on a low temperature level in the near-atmospheric pressure range.\textsuperscript{(2)} Numerous experimental DBD studies have explored effects on gas mixtures, electrical characteristics, and reactor geometries. Related work has focused on maximizing the ultraviolet radiation from excited molecules produced in DBD. In this experiment, physical and chemical parameters were varied systematically to understand how the reactor design and electric parameters affect the energy transfer and NO conversion characteristics in DBD process. Geometric parameters are the radius of discharge electrode (gap distance between electrodes), surface roughness of discharge electrodes, and others. Electrical and chemical parameters are peak voltage, initial concentrations, and residence time (flow rate). The light emission characteristics of DBD were investigated with a silicon PIN detector and a fiber optic spectrometer for the comparison of the relative discharge intensity.
according to the variation of physical parameters. Especially, emission characteristics in the wavelength of ultraviolet range were mainly investigated.

2. Experiments

2.1 Plasma Reactor
Most experiments were carried out at ambient temperature and pressure in a dielectric barrier discharge reactor. The reactor geometry was a concentric cylinder. Discharge electrodes (center electrodes) with different diameters were used, ranging from 15 mm to 35 mm (15, 20, 25, 30, and 35 mm). We obtained five annular gaps of a 14.5, 12, 9.5, 7, and 4.5 mm with those discharge electrodes. Each discharge electrode made of stainless steel had two types of surface, coarse and smooth. Cylindrical dielectric barriers made of pyrex was wrapped outside with a copper film tape which served as a grounded electrode. The inner diameter and thickness of the pyrex tube were 44 mm and 3 mm, respectively. The length of grounded copper film tape was 400 mm and the discharge plasma region could be reduced by limiting the length of the copper film. The end of the Pyrex tube was blocked with two teflon plates. Gas entered into the inlet of the reactor traveled vertically along the discharge electrode, and exited through the outlet. Instead of the outlet teflon cap, another cap with a viewport could be installed in the outlet to observe the microdischarge and to measure emission spectrum. A sketch of the plasma reactor is shown in figure 1.

![Fig. 1 Sketch of the dielectric barrier discharge reactor used in this experiment](image)

2.2 Experimental Setup

2.2.1 Power Supply
High voltage AC power was applied to the plasma reactor. The alternating voltage source was a high voltage transformer with a voltage adjuster that varies from 0 to 220 volts. The operating frequency was 60 Hz. We obtained the output voltages from 0 to 35 kV in experiments.

2.2.2 Power measurement
The experimental setup to obtain a charge-voltage plot on an oscilloscope is shown in figure 2. High voltage applied to the reactor was measured by using a Tektronix P6015A 1000X high voltage probe. The charge was determined by measuring the integrating capacitor of 1 µF with Tektronix P5100 100X voltage probe (2500 volt peak). The total current was obtained by differentiating these curves with a Tektronix oscilloscope (TDS 744A). Power consumption in the reactor (power consumed by the discharge plasma) was measured using the voltage and current traces recorded by the oscilloscope. The dissipated power was determined by multiplying and averaging the voltage and current, and also calculated from charge-voltage plot.

![Fig. 2 Experimental setup for obtaining Q-V plot](image)

2.2.3 Measurement of Light Emission
The experimental setup for the emission spectrum of DBD is shown in figure 3. A fiber optic irradiance probe was placed in front of the cap with a viewport. The probe sends signals to the spectrometer through the optical fiber. The spectrometer (S2000 Miniature Fiber Optic Spectrometer, Ocean Optics, Inc.) has a linear CCD-array silicon detector responsive from 320-1100 nm. Data obtained from the spectrometer were analyzed by operating the software OOIBase. It provided information on the relative irradiance of each wavelength. The light emission characteristics of DBD were also investigated with a silicon PIN detector for comparison of the relative discharge intensity according to the variation of physical parameters. The reactor was thoroughly covered with a curtain for measuring the light produced in the reactor only.
2.2.4 Measurement of NOx

A Thermo-Electron (model 42H) chemiluminescent NOx analyzer was used to measure nitrogen oxides in the gas stream entering and exiting from the reactor. Concentrations of NO were varied from 200 ppm to 400 ppm. Residence time was varied from 2 to 6 seconds.

3. Results and Discussions

The effect of the electrode diameter was studied on electrical characteristics and NOx conversion. The gap distance was varied directly as the diameter of discharge electrodes. We obtained five annular gaps of a 14.5 mm (35 mm dia.), 12 mm (30 mm dia.), 9.5 mm (25 mm dia.), 7 mm (20 mm dia.) and 4.5 mm (15 mm dia.). The peak voltage was changed from 14.8 kV to 34.8 kV.

3.1 Effect of Peak Voltage and Electric Field on Average Dissipation Energy

The effects of peak voltages and electric fields on average dissipation energy are shown in figure 4 and 5. Average dissipation energy was determined by multiplying and averaging the voltage and current, and it was also calculated from charge-voltage plot. In charge-voltage Lissajous figure, the area inside the parallelogram is equal to the energy dissipated during one cycle.(3) As shown in figure 4, the average dissipation energy increases as the peak voltage increases. The slope increased with decreasing the radius of discharge electrodes.

In the case of changing the radius of discharge electrodes, the curvature of electrode also varies. For the effect of the electrode shape, the maximum electric field on each discharge electrode was calculated. The relation between the electric field and the average dissipation energy is shown in figure 5. The average dissipation energy is proportional to the electric field. Each discharge electrode has two types of surface, coarse and smooth. As the diameter of discharge electrodes increased, the slope decreased. The slope of lines was not affected by the surface condition of discharge electrodes. The dissipation energy increased as the diameter of discharge electrodes decreased at the same electric field.

![Fig. 4 Effect of peak voltage on dissipation energy](image)

![Fig. 5 Effect of electric field on dissipation energy](image)

The corona onset voltage decreases as the gap distance decreases. When the driving voltage has reached a value $V_{cs}$ (corona onset voltage), the voltage across the gap has reached its sparking potential $V_s$, and breaks down into a corona. Because the gas in the gap becomes conductive with an impressed voltage of $V_{cs}$, the voltage drop across the gap remains essentially constant at $V_s$ with further increases in $V_{cs}$.(4) The sparking potential, $V_s$, is readily obtainable from charge-voltage plot. Figure 6 shows the effect of gap distance and electrode conditions (smooth or coarse) on the sparking potential. The sparking potential increased linearly as the gap distance and the sparking potential for the electrode of a coarse surface was a little bit lower than that for the electrode with a smooth.
3.2 Effect of Physical, Chemical, and Electrical Parameters on NOx Conversion Efficiency

Non-thermal plasmas give efficient means for selective partial oxidation of NO to NO₂. At a low energy density, the conversion ratio of NO to NO₂ was observed quantitatively. This process is due to the reaction of NO with ozone formed in discharges:

O₂ + (discharge) → 2O  
O + O₂ + M → O₃ + M  
NO + O₃ → O₂ + NO₂

Reaction 3 between ozone and NO is relatively fast ($k_3(298K) = 1.8 \times 10^{14}$ cm³ molecule⁻¹ s⁻¹). Usually, the increase of the energy deposition leads to a significant decrease in the concentrations of both NO and NO₂. However, in our experiments, most of NO was converted to NO₂. Figure 8 shows the NO conversion trend as energy density. Experiments were done at NO concentration of 200, 300, and 400 ppm. They are plotted as a function of electrical energy density (J/L), which is electrical power delivered to plasma divided by the gas flow rate. This experiment was carried out at room temperature with an inlet gas mixture of NO in air. As shown in figure 8, the concentration of NO decreased as energy density. As the concentration of NO decreased, that of NO₂ increased. The total concentration of NOx (sum of NO and NO₂) was not changed significantly. The concentration of NO dramatically dropped until 100 J/L, and about 75% of NO was converted to NO₂ at the energy density of 100 J/L and above. NO was converted to NO₂ rapidly as soon as the dielectric barrier discharge was produced and then NO was no more converted to NO₂ around 200 J/L. As one can see in the figure, the conversion rate saturates beyond 200 J/L.

Figure 9 shows the effect of the initial concentration on the conversion rate of NO. The initial concentration was varied from 200 ppm to 400 ppm. The diameter of discharge electrodes was varied from 20 mm to 35 mm (smooth and coarse surface) and the residence time of NO in a reactor was varied from 2.47 to 6 seconds. For 200 and 300 ppm cases, most of NO was converted to NO₂ at the energy density of 50 J/L. The conversion rate was over 99% at the energy density of 50 J/L and above. For 400 ppm case, the maximum conversion rate of NO was almost 80%. As shown in figure 9, as the initial concentration increased, the energy density needed to obtain the same conversion rate also increased. An experiment was carried out to determine the effect of residence time on the NO conversion. The residence time was varied from 2.47 to 6 seconds. The diameter of discharge electrodes (coarse and smooth surface) and the initial concentration of NO were varied from 25 mm to 35 mm and from 200 ppm to 400 ppm, respectively. One may note that the residence time affects the NO conversion rate. It was expected that the increase of the
residence time would improve the NO conversion rate. However, in our experimental range, the NO conversion rate was not much affected by the residence time as shown in figure 10.

Figure 11 shows the effect of the electrode surface condition on the NO conversion rate. The conversion rate for the smooth electrode was higher than that for the coarse electrode at the same energy density.

Figure 12 shows the effect of the electrode radius on the NO conversion. The experiment was carried out with an inlet gas mixture of 200, 300 and 400 ppm NO in air. The residence time and the diameter of discharge electrode were varied from 2.47 seconds to 6 seconds and from 25 mm to 35 mm, respectively. Every experiment was conducted with each type of discharge electrodes with smooth and coarse surfaces. As shown in figure 12, the NO conversion rate of the discharge electrode with 35 mm radius was higher than that of the discharge electrode with 30 mm radius at the same residence time and the same energy density.

3.3 Study on the Emission Characteristics of Dielectric Barrier Discharge

Light emission was observed inside the reactor during the dielectric barrier discharge. As already mentioned, the non-thermal plasma gives efficient means for the selective partial oxidation of NO to NO$_2$. However, for converting NOx to N$_2$ and O$_2$ effectively, catalysts are required. We are planning to treat NOx using a dielectric barrier discharge reactor combined with TiO$_2$ photocatalyst. The photocatalyst becomes a strong oxidizer or reducing agent to decompose pollutants under an ultraviolet light. Ozone absorbs lights in the short-ultraviolet wavelength region (200–300 nm), the Hartley band, with maximum absorption at 253.7 nm. The emission spectrum was measured with a spectrometer in air. It was to determine the wavelength of light emitted from the dielectric barrier discharge reactor. Figure 13 shows typical emission spectrum. The wavelengths were measured from 335 nm to 800 nm. Relatively high peaks were appeared around wavelengths of 357, 375, 380, 394, 400 and 405 nm. As the dissipation energy increased, the magnitude of peak also increased.

Figure 14 shows the effect of the dissipation energy in a DBD reactor on the emission intensity. The relative intensity means the relative magnitude of peaks. As shown in figure 14, the relative intensity increased as the
dissipation energy (or peak voltage). Figure 14 also shows the effect of the residence time on emission characteristics. As the flow rate increased, the relative intensity decreased. The result was caused by the effect of the ozone concentration. Ozone absorbs light in the short-ultraviolet wavelength region. The wavelength of 380 nm is not in a short-ultraviolet wavelength region and the ozone absorption of this is relatively low. However, some of light is absorbed by ozone. The decrease of the flow rate is equivalent to the residence time increases, and thus the accumulation of ozone will increase in the reactor and the absorption of light will increase, too.

![Fig. 13 Typical emission spectrum of dielectric barrier discharge in the 235–500 nm range.](image)

![Fig. 14 Effect of dissipation energy and residence time on emission intensity (electrode diameter: 35(S), wavelength: 380 nm)](image)

4. Conclusions

The experiments have resulted in the following conclusions;

- The average dissipation energy was proportional to the peak voltage and the electric field. The sparking potential increased linearly with the decrease of the radius of the discharge electrode.
- The sparking potential for the coarse electrode was lower than that for the smooth electrode.
- The optimum gap distances for the highest dissipation energy existed in a given voltage.
- NO was converted to NO₂ rapidly at a low energy density, and the energy density needed to obtain the same conversion rate increased as the initial concentration.
- The NO conversion rate was affected by the surface conditions and the radius of the discharge electrodes.
- The magnitude of emission spectrum increased as the dissipation energy and the flow rate.

Results presented in this paper show that geometric, chemical, and electrical parameters should be considered for optimum reactor design, and further investigations are required.

References


