Gas-Phase Removal of NO from Gas Streams via Dielectric Barrier Discharges

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■ The removal of NO from gas streams using a plasma generated by dielectric barrier discharges is investigated experimentally. NO removal efficiencies of >95% are achieved. The removal efficiency of NO is measured as a function of NO, O_2 , CO_2 , and H_2O_g concentrations, as well as temperature of the gas stream. Removal of NO decreases under dry conditions with increasing O_2 concentration at ≈ 24 °C due to plasma oxidation of N_2 . At elevated temperatures, increasing H_2O_g concentration and decreasing CO_2 concentration result in increasing NO removal efficiencies. NO removal increases with increasing H_2O_g concentration due to the increase in production of OH. CO_2 causes a decrease in the amount of energy deposited into the gas stream, resulting in lower NO removal efficiencies. The overall results suggest that dielectric barrier discharges have the potential as an alternative to existing postcombustion technology for the removal of NO from gas streams.

Introduction

Nitric oxide, a gaseous air contaminant, is harmful to human health when inhaled, damages vegetation, and degrades materials through acid deposition (1, 2). Once released into the atmosphere, NO can be oxidized to form nitrogen dioxide, which is also harmful to humans and can corrode materials (3). When illuminated by sunlight, atmospheric NO reacts with gaseous hydrocarbons and O_2 to form photochemical oxidants, such as peroxyacetal nitrate (PAN) and ozone (4). Emission of NO into the atmosphere may be responsible for the inability of large metropolitan areas to meet the National Ambient Air Quality Standards for O_3 (5). Exhaust gases from the combustion of fossil fuels such as oil and coal are one of the major sources of anthropogenic NO. In 1985, $18.6 \times$ 10^6 metric tons of NO_x was released into the atmosphere by human activities in the United States (6). Stationary point sources such as electric utilities and other combustion processes contribute $\approx 10.6 \times 10^6$ metric tons of NO_r annually, which accounts for 57% of the total anthropogenic emissions from the United States for 1985 (6). About 90-95% of NO_x emitted from combustion sources exists initially as NO but forms other oxides of nitrogen in the atmosphere. As a result, minimizing the emissions of NO from these stationary sources would reduce the concentration of NO_x in the atmosphere and therefore reduce its adverse effects.

Two mechanisms that are primarily responsible for NO_x formation during combustion are thermal NO_x and fuel NO_x . Thermal NO_x results from oxidation of N_2 contained in the combustion air, whereas fuel NO_x originates from the oxidation of chemically bound nitrogen in the fuel. Abatement of NO emissions can be achieved by (1) modifying the combustion process, such as by use of low- NO_x burners, gas recirculation, and staged combustion, and (2) using postcombustion removal methods. Combustion

[§] Present address: Institute of Environmental Engineering, National Central University, Chungli, Taiwan 32054, Republic of China. modification strategies have been widely used to reduce the emissions of NO_x ; however to meet current emission standards for NO_x , postcombustion removal techniques must also be evaluated (7). Selective catalytic reduction (SCR) is the most advanced technology currently used to remove NO from gas streams on a postcombustion basis. Particulate material and SO₂ commonly found in flue gas streams generated by the combustion of fossil fuel tend to poison catalysts and, therefore, lower SCR performance (8).

Another postcombustion removal method for NO_x is selective noncatalytic reduction (SNCR). SNCR relies on the injection of ammonia into the gas stream to chemically reduce NO to N_2 and H_2O in the presence of O_2 . However, SNCR is effective only between 900 and 1100 °C (9) and NH₃ may not be totally consumed in the process, thereby leading to the release of NH₃ from the device to the atmosphere (8). Limitations of these existing control technologies have motivated research to develop new methods to remove NO from gas streams.

One alternative to SCR and SNCR processes that has been studied since the 1970s is gas-phase plasma oxidation of NO with electron beams (10). The basis of this method is to initially generate gas-phase radicals, such as O, OH, and HO_2 by electron impact excitation and ionization. Subsequent reactions with those radicals then oxidize NO to form HNO_3 . A suggested reaction scheme is shown with eqs 1–5 (11, 12). The resulting HNO_3 can then be chem-

rate coefficient

$2NO + O_2 \rightarrow 2NO_2$	$5.0 \times 10^{-38} \text{ cm}^6/\text{s}$	(1)
NO + O + M \rightarrow NO ₂ + M	$7.0 \times 10^{-32} \text{ cm}^6/\text{s}$	(2)
NO + O ₃ \rightarrow NO ₂ + O ₂	$1.6 \times 10^{-14} \text{ cm}^3/\text{s}$	(3)
NO + HO ₂ \rightarrow NO ₂ + OH	$6.6 \times 10^{-12} \text{ cm}^3/\text{s}$	(4)
$NO + HO_2 \rightarrow NO_2 + OH$	$6.6 \times 10^{-12} \text{ cm}^3/\text{s}$	(4)
$NO_2 + OH \rightarrow HNO_3$	$1.1 \times 10^{-11} \text{ cm}^3/\text{s}$	(5)

ically neutralized to NH_4NO_3 by injection of NH_3 or neutralized to $Ca(NO_3)_2$ by injection of $Ca(OH)_2$ into the gas stream. The resulting particles can then be removed from the gas stream by a particle control device (13). Although plasma oxidation using electron beams is effective at removing NO from gas streams, its high cost and potential X-ray hazard have motivated research into other methods to generate these gas-phase radicals (14, 15).

In this study, the dielectric barrier discharge (DBD) is experimentally evaluated as an alternative method to generate gas-phase radicals with plasmas. DBDs have been used to generate atmospheric pressure plasmas that have been employed as ozonators. Our goal is to remove NO from simulated gas streams. In DBDs, the plasma is sustained between one or more dielectric surfaces backed by a conductor. A repetitively pulsed or ac voltage is applied to the electrodes, resulting in a filamentary pulse periodic plasma. Gas flowing through a DBD is processed by many such discharge pulses. Excitation of atmospheric pressure plasmas in this fashion is quite efficient because arcs are prevented by the short duration of the individual current pulses, and the electron temperature of the plasma is well matched to the electronic and dissociative states of molecular gases.

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Figure 1. Schematic of the gas generation system, dielectric barrier discharge reactor, and gas sampling and detection system used for elevated-temperature experiments. The experimental system used for room-temperature experiments was the same except for the H₂O_g generation system and the heat exchanger located around the reactor.

In the following sections, the laboratory-scale DBD reactor used in this study is described followed by our experimental results and a discussion of the effectiveness of DBDs to remove NO from gas streams. This is the first time that DBDs have been experimentally evaluated as a means of removing NO from simulated gas streams.

Experimental Apparatus

The experimental apparatus used in this study is schematically shown in Figure 1 and includes a continuous-flow gas generation system, laboratory-scale DBD reactor, and gas sampling and detection system. The gas generation system provides a gas stream containing N₂, O₂, CO₂, NO, and H_2O_p with a specific composition, mass flow rate, and temperature. Dry-grade N2, dry-grade air, CO2, and NO from compressed gas cylinders were regulated by mass flow controllers (Tylan Inc., Model No. FC-260). For the room-temperature experiments ($T \approx 24$ °C), the concentration of H_2O_g ([H_2O_g]) was controlled upstream of the reactor by passing a portion of the dry gas stream through a controlled-temperature H_2O_ℓ bath before the gas stream was mixed with the balance of the carrier gas and NO. $[H_2O_g]$ was determined with a dew point hygrometer (General Eastern, Model No. Hygro-M1) and a Type K thermocouple (Omega, Model No. CASS-18). For the elevated-temperature experiments, the gas stream was initially heated to ≈ 350 °C with an electrical resistance heater (Lindberg, Type No. 59344) to ensure complete evaporation of H_2O_ℓ droplets that were injected into the gas stream with a peristaltic pump (Gilson, Model Miniplus 2) and a hypodermic needle (gauge No. 20). The resulting $[H_2O_g]$ was controlled by regulating the volume feed rate of H_2O_ℓ injected into the high-temperature gas stream. The gas stream was kept at ambient pressure in the DBD reactor for all tests.

The cylindrical DBD reactor consisted of a crystalquartz tube with an inner diameter of 4 cm and a wall thickness of 0.2 cm (Figure 1). One of the coaxial elec-

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trodes was made of 0.24-cm-diameter tungsten rod and the other electrode was a stainless steel wire mesh screen. The rod electrode was aligned vertically along the center line of the reactor and the wire mesh electrode was wrapped around the outside of the reactor. A glass ball with a diameter of 0.4 cm was used to cover the end of the rod electrode to inhibit the formation of electrical discharges at the end of the electrode. Near-isothermal conditions $(\pm 5 \text{ °C})$ were obtained within the discharge volume during the elevated-temperature experiments by forcing hot air through an annular quartz tube air jacket having an outside diameter of 7.5 cm. Plasma was sustained within a volume of 350 cm³ by a 60-Hz ac high-voltage power supply (Variac, Model No. WT10MT3 and MagneTek Jefferson Electric, Model No. 721-411). $[NO_x]$ was monitored with a chemiluminescence NO_x analyzer (Monitor Laboratory, Model 8840HL) after dilution with dry-grade N_2 (Monitor Laboratory, Model 8730) to cool and reduce the $[H_2O_g]$ and $[NO_x]$ in the gas stream before detection. The detector was calibrated by injecting gases having known NO concentrations upstream of the gas dilutor.

Unless specified, inlet [NO] was controlled at ≈ 250 ppmv (±8 ppmv). The gas flow rate was controlled at 3 and 2.7 standard L/min for room-temperature and elevated-temperature experiments, respectively. This condition corresponds to a gas residence time of 5.3 s in the plasma for a gas stream at 130 °C. Initial conditions for each test were recorded after the system reached steady-state conditions. Voltage from the power supply was then increased in a stepwise manner to values between 21 and 25 kV (peak value). [NO] was then recorded after it stabilized at its new value. The reactor's power supply was then shut off and the system was monitored to ensure that the inlet conditions had not changed during the test. The removal efficiency of NO (η_{NO}) achieved by plasma processing is specified by eq 6. The subscripts in eq 6 denote

$$\eta_{\rm NO} (\%) = \frac{[\rm NO_{(off)}] - [\rm NO_{(on)}]}{[\rm NO_{(off)}]} (100\%)$$
(6)



Figure 2. Dependence of η_{NO} on inlet $[O_2]$ and applied voltage for dry gas streams. The dielectric barrier discharge reactor was at \approx 24 °C, $[NO] \approx 250$ ppmv, $[H_2O_g] < 3$ ppmv, $[O_2]$ as specified, and N_2 as the carrier gas. η_{NO} decreases with increasing $[O_2]$ due to increased oxidation of N_2 . Error bars indicate one standard deviation from four sets of measurements.

whether the power supplied to the DBD cell was turned on or off.

Results and Discussion

The dependence of η_{NO} on inlet $[O_2]$ in a dry gas stream is shown in Figure 2. The inlet gas temperature was 24 °C. The [NO] was 250 ppmv and the balance is N_2 . η_{NO} gradually increases as inlet $[O_2]$ increases from 0% to 20% by volume. In the absence of O_2 , NO was most likely chemically reduced to N_2 by N atoms as described by reactions 7 and 8 (11). N atoms were generated by elec-

rate coefficient

$$\begin{array}{ll} {\rm NO} + {\rm N}({}^4{\rm S}) \rightarrow {\rm N}_2 + {\rm O}({}^3{\rm P}) & 2.2 \times 10^{-11} \ {\rm cm}^3/{\rm s} & (7) \\ {\rm NO} + {\rm N}({}^2{\rm D}) \rightarrow {\rm N}_2 + {\rm O}({}^3{\rm P}) & 1.8 \times 10^{-10} \ {\rm cm}^3/{\rm s} & (8) \end{array}$$

tron impact dissociation of N_2 and dissociative recombination of N_2^+ . As $[O_2]$ was increased in the reactor, N atoms generated within the plasma not only reduce NO through reactions 7 and 8, but also form NO through reactions 9 and 10 (11). Generation of NO is also possible

rate coefficient

$N + O_2 \rightarrow NO + O$	$1.0 \times 10^{-16} \text{ cm}^3/\text{s}$	(9)
$N + O_3 \rightarrow NO + O_2$	$3.7 \times 10^{-13} \text{ cm}^{3}/\text{s}$	(10)
$NO_2 + O \rightarrow NO + O_2$	$5.0 imes 10^{-12} \text{ cm}^3/\text{s}$	(11)

by chemical reaction of NO₂ with O as indicated by eq 11. O atoms are produced by electron impact dissociation of O₂ and excitation transfer from electronically excited N₂ [i.e., N₂(A) + O₂ \rightarrow 2O + N₂]. O₃ is also generated by reaction of O atoms and O₂. O and O₃ in the DBD reactor (16) can also oxidize NO to form NO₂ through reactions 2 and 3.

The overall gas-phase removal mechanisms for NO are complicated for a gas stream containing both O_2 and N_2 since NO can simultaneously be generated and removed (17). For a dry gas stream containing 250 ppmv NO and a low $[O_2]$, the rate of removal of NO dominates the rate of generation of NO, resulting in its high rate of removal. As $[O_2]$ increases, the rate of generation of NO increases more rapidly than the rate of NO removal, and this leads to lower values of η_{NO} . The rates of removal and generation of NO almost balance each other as $[O_2]$ increases to 20% by volume, resulting in low values of η_{NO} ($\leq 2\%$). η_{NO} increases with increasing applied voltage in all cases due to increasing power deposition. The operational range used



Figure 3. η_{NO} as a function of $[O_2]$ and voltage with the existence of H_2O_g . Experiments were performed at ≈ 24 °C with 250 ppmv NO, $[H_2O_g] = 2.6\%$ by volume, and N_2 as the carrier gas. Local maximum values for η_{NO} occur at $[H_2O_g] \approx 5\%$ by volume.

here, 21-25 kV, was a consequence of physical limitations of our particular device. Higher removal of NO is expected in all cases by operating the power supply at a higher voltage or a higher frequency.

The dependence of $\eta_{\rm NO}$ on inlet $[O_2]$ with 2.6% by volume $[H_2O_g]$ is shown in Figure 3. $\eta_{\rm NO}$ increases from 18% to a local maximum value of $\approx 48\%$ as $[O_2]$ increases to $\approx 5\%$ by volume at 25 kV. As $[O_2]$ increases above 5% by volume, $\eta_{\rm NO}$ decreases to $\approx 38\%$ at 25 kV. In the absence of O_2 , generation of gas-phase radicals is low (16), resulting in low $\eta_{\rm NO}$. As $[O_2]$ increases, the rate of generation of OH also increases, primarily by $O(^1D)$ atom abstraction of H from H_2O_g as shown by reactions 12 and 13, where the rate coefficient for (12) depends on a reduced electric field (E/N).

rate coefficient

$$\begin{array}{ll} {\rm e} + {\rm O}_2 \rightarrow {\rm O} + {\rm O}({}^1{\rm D}) & (12) \\ {\rm O}({}^1{\rm D}) + {\rm H}_2 {\rm O}_{\rm g} \rightarrow 2{\rm OH} & 2.2 \times 10^{-10} \ {\rm cm}^3/{\rm s} & (13) \end{array}$$

Increasing the inlet $[O_2]$ also results in a higher rate of generation of O and O_3 . From the perspective of removing NO from the gas stream, O and O_3 can oxidize NO to form NO_2 , which can then be further oxidized by OH to form HNO_3 . As a result, O and O_3 can enhance NO removal in humid gas streams due to the accompanying generation of OH. From the perspective of forming NO, O atoms and O_3 molecules may also react with N_2 and NO_2 to generate NO as shown in reactions 10 and 11. Decreasing values for $\eta_{\rm NO}$ with increasing $[O_2]$ for $[O_2] > 5\%$ by volume indicate that the rate of formation of NO begins to approach the rate of removal of NO via oxidation with OH at 2.6% by volume H_2O_g . These results imply that there is an optimum $[O_2]$ for a specific inlet gas composition which maximizes NO removal. Such observation is in agreement with results obtained by Menegozzi and Feldman (18). $\eta_{\rm NO}$ also increases with increasing values of $[H_2O_g]$, as shown in Figure 4. The increase in η_{NO} is principally attributed to the increased rate of generation of OH and HO_2 with increasing $[H_2O_g]$, since $[O_2]$ remains constant (16).

We also investigated the dependence of η_{NO} on $[CO_2]$, $[H_2O_g]$, $[O_2]$, [NO], and $[N_2]$ at elevated temperatures since the gas streams typically emitted from fossil fuel combustors contain these gases. Results were obtained for gas streams containing 250 ppmv NO at 70, 100, 130, and 160 °C, and an applied voltage of 25 kV (Figure 5). As $[H_2O_g]$ increases from 0% (\leq 3 ppmv) to 3% by volume, η_{NO} increases from 20% to 75% with little dependence on tem-



Figure 4. Dependence of η_{NO} on inlet $[H_2O_g]$ and voltage for gas streams at ≈ 24 °C containing 250 ppmv NO, 20% by volume O_{2^*} , $[H_2O_g]$ as specified, and N_2 as the carrier gas. Increase in $[H_2O_g]$ results in the generation of more OH and HO₂ and a larger rate of oxidation of NO to HNO₃. Error bars indicate one standard deviation from four sets of measurements.



Figure 5. η_{NO} as a function of inlet $[H_2O_g]$ at specified dry-bulb temperatures and at 25 kV for gas streams containing 250 ppmv NO, 6% by volume O_2 , 12% by volume CO_2 , and N_2 as the carrier gas. Increasing $[H_2O_g] > 5\%$ by volume can either increase or decrease η_{NO} depending on the minimum required E/N value.

perature. However, as $[H_2O_g]$ increases from 3% to 15% by volume at 130 and 160 °C, $\eta_{\rm NO}$ increases to values of >95%. The increase in $\eta_{\rm NO}$ with increasing $[H_2O_g]$ is a result of generating more OH and HO₂ by electron impact dissociation of H_2O_g and excitation-transfer reactions. These radicals then oxidize NO to form HNO₃, which is the desired product that can be more readily removed from gas streams than NO.

The dependence of η_{NO} on gas temperature and gas composition is complicated because of the impact of electron impact excitation rates on $\eta_{\rm NO}$. As gas temperature decreases, the gas density, N, increases, and E/Ndecreases at a constant applied potential. Since the electron energy distribution function depends on E/N for a specific gas composition, lower values of E/N result in lower average electron energies, which reduce the rate of generation of OH, O, and HO_2 . O_2 , H_2O_g , and CO_2 are electronegative gases, whereas N_2 is not (15, 19). Therefore, increasing $[O_2]$, $[H_2O]$, or $[CO_2]$ of the gas stream increases the rate at which electrons attach or are removed from the gas stream. The result is that the minimum E/Nrequired to initiate and sustain the plasma, $(E/N)_{o}$, increases. Since the voltage applied to the DBD is sinusoidal, the plasma can be sustained for only that fraction of the cycle for which the applied $|E/N| > (E/N)_{o}$. Since $(E/N)_{o}$ increases as $[O_2]$, $[H_2O_g]$, or $[CO_2]$ is increased, the fraction of the cycle over which the plasma can be sustained de-



Figure 6. Dependence of $\eta_{\rm NO}$ on inlet $[\rm CO_2]$ and voltage for gas streams at 130 °C with 400 ppmv NO, 6% O₂ by volume, 6% H₂O_g by volume, and N₂ as the carrier gas. The decreasing values for $\eta_{\rm NO}$ are likely caused by the electronegative nature of CO₂, which decreases power deposition.



Figure 7. Percent (solid line) and absolute (dashed line) removal of NO as a function of inlet [NO] and voltage at 130 °C for gas streams with $O_2/H_2O_g/CO_2 = 6\%/6\%/12\%$ by volume and N_2 as the carrier gas. At a constant power deposition, absolute removal of NO continues to increase while percent removal decreases with increasing inlet [NO].

creases, as does the average power deposition. A systematic decrease in the value of $\eta_{\rm NO}$ as the concentrations of these electronegative gases increase may in part be explained by a decrease in power deposition due to the increase in $(E/N)_{\rm o}$. Such behavior is observed for $\eta_{\rm NO}$ at gas temperatures of 70 and 100 °C, as indicated in Figure 5.

The dependence of η_{NO} on inlet $[CO_2]$ is shown in Figure 6 for gas streams containing 400 ppmv NO. η_{NO} decreases with increasing $[CO_2]$ up to 12% by volume. If dilution of $[N_2]$ by CO_2 was the only effect caused by increasing $[CO_2]$, then η_{NO} should increase as a result of the decreased formation of NO by oxidation of N_2 . CO_2 , however, is an electronegative gas that acts to increase the self-sustaining voltage of the gas stream. In these cases, the decrease in power deposition resulting from the increase in $(E/N)_o$ dominates over the effects of dilution.

 $\eta_{\rm NO}$ for gas streams with inlet NO concentrations ranging from 150 to 500 ppmv, which are typical values for gases generated by the combustion of coal, are shown in Figure 7. $\eta_{\rm NO} > 94\%$ was achieved for an inlet [NO] of 150 ppmv. Although the percent removal of NO decreases with increasing inlet [NO], the absolute removal increases with increasing [NO]. Commercial ozonizers can achieve increased power deposition using similar technology by operating at higher frequencies or higher E/N values. Under these conditions, the percentage removal of NO should not have as strong a dependence on [NO] as shown here.

 $[NO_x]$ and η_{NO_x} were also monitored for tests at elevated temperatures. When [NO] and $[O_2]$ were at 250 ppmv and 6% by volume, respectively, $[NO_x]$ was ≈ 285 ppmv, which

indicates that some NO had been oxidized by the O₂ in the gas stream to form NO₂ and had been detected as NO_x. Values for $\eta_{\rm NO_x}$ were very similar to $\eta_{\rm NO}$ except the values were 10–15% lower than $\eta_{\rm NO}$. Slightly lower values for $\eta_{\rm NO_x}$ implies that most NO is oxidized to HNO₃, but a small portion (\approx 5–10%) of NO is oxidized to NO₂.

Numerous attempts have been made to characterize the power deposited into the DBD reactor with the method described by Carlins (20). Accurate measurement of the power deposited into the gas stream is difficult because of the numerous and very short time response pulses (up to tens of nanoseconds) of current that occur when the power supply's voltage exceeds the breakdown voltage. Also, the high-voltage transformer used as part of the power supply is internally grounded at the middle of the secondary winding (center tap). Such grounding makes it difficult to simultaneously measure the output of the power supply's voltage and current. Results from experimental measurements and numerical modeling estimate that tens of millijoules per milliliter of gas is required to achieve at least 90% removal of NO from a gas stream containing 250 ppmv NO at 130 °C and 1 atm (16).

A full-scale DBD reactor used at a typical power plant that combusts fossil fuel would operate at temperatures, pressures, compositions of gas streams, and gas residence times similar to conditions that were simulated in this study. The laboratory measurements suggest that such conditions would not cause significant problems when DBD technology is used at a typical power plant. Also, the actual power supply for the DBD is much simpler than those currently used with electrostatic precipitators. The actual system could consist of DBD reactors, located in parallel, similar to a tubular-type electrostatic precipitator. In addition, DBD technology is fairly mature as a result of its use to generate O_3 . Although there are many characteristics in favor of DBD technology, there are still numerous aspects of DBD technology that need to be explored.

Further research is needed to develop a commercially available DBD reactor that removes NO from gas streams. Future research directions include (1) characterization of the influence of dispersed particulate material that exists in typical flue gas streams on DBDs, (2) accurate measurement of the power deposited into the DBD reactor, (3) optimization of a power supply that operates at higher frequencies, and (4) optimization of the geometry of the DBD reactor.

Conclusions

Gas-phase removal of NO with dielectric barrier discharges has been experimentally investigated with a laboratory-scale reactor and simulated flue gas streams. Experimental results indicate that 95% removal of NO can be achieved for conditions that simulate the composition and temperature of gases generated by the combustion of fossil fuel. NO removal efficiency is sensitive to gas temperature, gas composition, and power deposition. In general, increasing $[H_2O_g]$ increases the removal of NO, while increasing $[CO_2]$ decreases the removal efficiency of NO. There is also an optimal concentration of O_2 to maximize the removal of NO at a specific concentration of H_2O_g . The use of DBDs has the potential as an alternative to existing postcombustion technology for removal of NO from gas streams.

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