Removal of SO₂ and NO from Gas Streams with Combined Plasma Photolysis
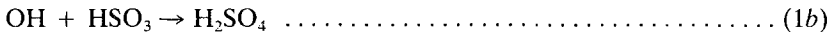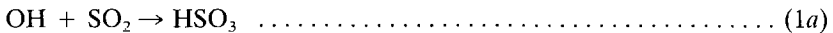
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ABSTRACT: Combined plasma photolysis (CPP) has been developed and experimentally demonstrated as a new method to simultaneously remove SO_2 and NO from gas streams. This laboratory-scale device integrates the use of a dielectric barrier discharge (DBD) plasma and ultraviolet radiation. The composition, temperature, and pressure of the treated gas streams simulate gases typically generated by the combustion of fossil fuels. Simultaneous removal efficiencies for SO_2 and NO are as high as 29% and 79%, respectively. CPP enhances SO_2 removal efficiencies by 25% when compared to using DBDs only. NO removal efficiency achieved by CPP is 9% less than NO removal efficiency achieved by DBDs only. SO_2 and NO removal efficiencies are limited by the power deposited into the gas stream that could be achieved with the existing power supply. This new device shows promise as a new technique to simultaneously remove SO_2 and NO from gas streams generated by the combustion of fossil fuels.
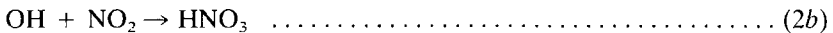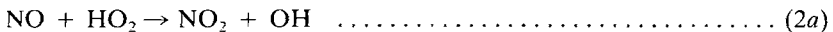
INTRODUCTION

Concerns over the adverse effects of atmospheric SO_2 and NO_x (NO and NO_2) on human health and the environment have resulted in more stringent standards for emissions of SO_2 and NO into the atmosphere from fossil-fuel-fired utilities as indicated by the Clean Air Act Amendments of 1990. In view of the constraints of air-quality-control technologies that currently remove either SO_2 or NO from gas streams (Lyon 1987; Meyler 1981; Rhoads 1990), it would be beneficial to develop a process that simultaneously removes SO_2 and NO from gas streams.

The possibility of applying gas-phase oxidation processes to simultaneously remove SO_2 and NO from flue gas streams has been studied during the previous two decades. The principle of this technology is based on the use of plasmas to generate gas-phase radicals, such as OH (hydroxyl), HO_2 (hydroperoxyl), and O (atomic oxygen). These gas-phase radicals oxidize SO_2 and NO in the gas stream to form H_2SO_4 and HNO_3, respectively. The primary reactions are presented below.

$$OH + SO_2 \rightarrow HSO_3 \quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad (1a)$$

$$OH + HSO_3 \rightarrow H_2SO_4 \quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad (1b)$$

and

$$NO + HO_2 \rightarrow NO_2 + OH \quad\quad\quad\quad\quad\quad\quad\quad\quad\quad (2a)$$

$$OH + NO_2 \rightarrow HNO_3 \quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad (2b)$$

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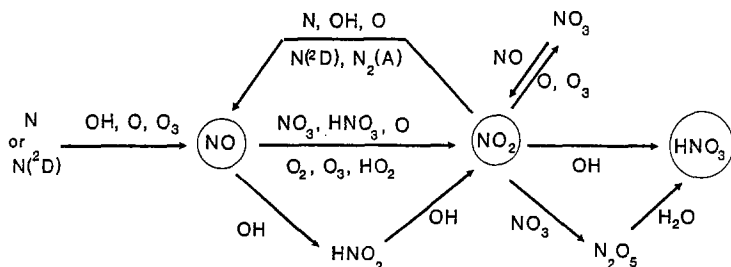
FIG. 1. Dominant Pathways to Oxidize NO to HNO₃ (Balbach 1991)

Dominant pathways to oxidize NO to HNO_3 are complicated as described by Fig. 1. H_2SO_4 and HNO_3 can then be chemically neutralized and removed from the gas stream by other more conventional aerosol particle removal devices (Paur et al. 1988).

Electron-beam-produced plasmas, an example of a gas-phase oxidation process, coupled with $NH_{3(g)}$ injection, can simultaneously remove SO_2 and NO from gas streams (Jordan 1988; Fuchs et al. 1988). However, high cost and potential X-ray hazards make this process economically infeasible to remove SO_2 and NO from gas streams generated by fossil-fuel-fired combustors (Jordan 1988). Therefore, it would be beneficial to develop other gas-phase oxidation processes that can simultaneously remove SO_2 and NO from gas streams.

The present paper describes and demonstrates a new gas-phase oxidation process that simultaneously removes SO_2 and NO from gas streams. This method, called combined plasma photolysis (CPP), uses dielectric barrier discharges (DBDs), also known as silent discharges, and ultraviolet (UV) radiation to generate gas-phase radicals, thereby allowing the simultaneous oxidation of SO_2 to H_2SO_4 and NO to HNO_3 (Chang et al. 1991). The resulting compounds can then be chemically neutralized with $NH_{3(g)}$ and removed from the gas stream by an aerosol particle removal device, such as a fabric filter or electrostatic precipitator.

COMBINED PLASMA PHOTOLYSIS

CPP integrates DBDs with UV photolysis to enhance the generation of gas-phase radicals in the gas stream beyond that possible with DBDs only. DBD is a gas discharge device that operates stably at atmospheric pressure. Moderate power deposition to the gas stream can be obtained with DBDs, and are used to generate O_3 for numerous industrial applications (Eliasson et al. 1987; Horvath et al. 1985). DBDs have also been studied as a possible technology to remove SO_2 and NO from gas streams (Browne and Stone 1985; Sardja and Dhali 1990; Chang et al., 1991, 1992a, 1992b; Chang 1992). Simultaneous removal efficiencies of SO_2 and NO have been demonstrated to be as high as 32% and 95%, respectively, with DBD (Chang et al. 1992b). In this past work, power generated and then deposited into the gas stream by the existing power supply limited the achievable removal efficiencies of SO_2 and NO from the gas stream.

DBDs sustained in typical flue gas streams generate O_3 as well as gas-phase radicals such as OH, HO_2, and O. The rate of generation of O_3 depends upon power deposited into the gas stream, composition of the gas

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stream—such as concentrations of O_2 ($[O_2]$) and $H_2O_{(g)}$ ($[H_2O_{(g)}]$)—and temperature of the gas stream (Carlins 1982).

O_3 can oxidize NO to form NO_2 (Willis and Boyd 1976), and it is well known that O_3 absorbs UV radiation with wavelengths ranging from 200 to 300 nm (Griggs 1968). Absorption of UV radiation by O_3 in the stratosphere limits the flux of UV radiation into the troposphere (Finlayson-Pitts and Pitts 1986). Photolysis of O_3 results in the production of $O(^1D)$ if the wavelength of incident radiation is less than 310 nm (Fairchild and Lee 1978) as described by (3). The product of the absorption

$$O_3 + h\nu(\lambda < 310 \text{ nm}) \rightarrow O(^1D) + O_2 \dotfill (3)$$
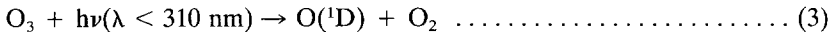
cross section and yield of $O(^1D)$ maximizes near 254 nm, the resonance line of Hg. Once produced by photolysis of O_3, $O(^1D)$ can then react with $H_2O_{(g)}$ in the gas stream to form additional OH radicals (Baulch et al. 1982).

$$O(^1D) + H_2O_{(g)} \rightarrow 2 \text{ OH} \dotfill (4)$$
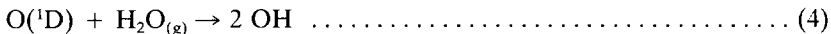
The additional OH radicals generated by UV photolysis of O_3 can then react with SO_2 and NO_x in the gas stream (Calvert and Stockwell 1984) as shown in (1) and (2). Therefore, simultaneous application of DBDs and UV radiation to a gas stream may enhance the removal efficiencies of SO_2 and NO achievable by DBDs only and is evaluated by the results presented in this paper.

EXPERIMENTAL DESIGN

An experimental apparatus was designed and constructed to evaluate the effectiveness of CPP to remove SO_2 and NO from simulated flue gas streams (Fig. 2). The apparatus was described in detail by Chang et al. (1992a), but will be described briefly here for clarity. The system consists of a continuous-
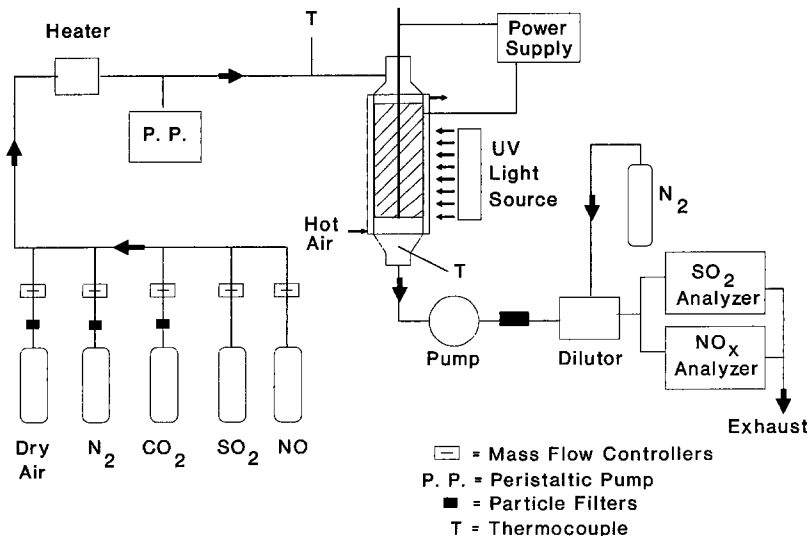

FIG. 2. Gas Generation System, Dielectric Barrier Discharge Reactor, UV Radiator, and Gas Sampling and Detection Systems

flow gas-generation system, laboratory-scale CPP reactor, and gas sampling and detection system. A gas stream with known mass-flow rates of O_2, N_2, and CO_2 were generated with compressed gas cylinders and mass-flow controllers. The resulting gas stream was then heated to 350°C with an electrical resistance heater to ensure complete evaporation of $H_2O_{(l)}$ droplets that were injected into the gas stream downstream of the heater with a peristaltic pump and hypodermic needle. Complete evaporation was confirmed by measurement of the $H_2O_{(l)}$ injected into the gas streams and measurement of the gas flow rate and $[H_2O_{(g)}]$ downstream of the injector. SO_2 and NO were then injected into the heated and humidified gas stream resulting in a gas stream with known composition, mass-flow rate, temperature, and pressure. All the tests were completed at 101 kPa (1 atm).

The DBD reactor was made of a crystal-quartz tube with an inner diameter of 4 cm and a wall thickness of 0.2 cm. The inner electrode, made of a 0.24 cm diameter tungsten rod, was aligned vertically along the centerline of the reactor. The outer electrode, made of a Type 304 stainless-steel wire mesh, was wrapped around the outside of the reactor. Hot air passed through an outer annular quartz tube to keep the reactor at 130 ± 5°C. Plasma was sustained within a volume of 350 cm³. Power aplied to the reactor was generated by a 60 Hz ac power supply at voltages between 23 kV and 25 kV (peak values). A medium-pressure mercury vapor lamp (Hanovia Inc., Newark, N.J., Model No. 6506A431) was used as the UV light source to radiate the reactor while generating DBDs.

Upon exiting from the reactor, the gas stream passed through stainless-steel tubing that was heated and insulated to prevent condensation of $H_2O_{(g)}$ in the gas-sampling lines. Just before detection of the gas stream, it was diluted with dry-grade N_2. N_2 injection was used to cool and reduce the $[H_2O_{(g)}]$, $[SO_2]$, [NO], and $[NO_x]$ of the gas stream before detection.

$[SO_2]$ was monitored with a fluorescence detection analyzer (Monitor Laboratory, San Diego, Calif, Model 8850HL). [NO] and $[NO_x]$ were monitored with a chemiluminescence detection analyzer (Monitor Laboratory, San Diego,Calif., Model 8840HL). O_3 generated with the DBDs was measured with a UV photometric analyzer (Thermo Environmental Instruments Inc., Franklin, Mass., Model 49).

Initial operating conditions were recorded when the system reached steady-state conditions with no power applied to the DBD reactor or the UV light source. Power was then applied to the DBD reactor and/or irradiated with UV light. $[SO_2]$, [NO], and $[NO_x]$ were then recorded when they stabilized at new values. The power applied to the DBD reactor and/or UV radiator was then shut off and the system was then monitored to ensure its return to initial conditions. Removal efficiencies of SO_2 (η_{SO_2}), NO (η_{NO}), and NO_x (η_{NO_x}) were then determined with the resulting measured values of $[SO_2]$, [NO], and $[NO_x]$, respectively. For example, η_{SO_2} was determined with

$$\eta_{SO_2} = \frac{[SO_2]_{off} - [SO_2]_{on}}{[SO_2]_{off}} \quad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots (5)$$

where the subscripts denote if the power supplies for the DBD reactor and/or UV radiator were on or off.

RESULTS AND ANALYSIS

The ability of the DBD reactor to generate O_3 was characterized to determine the applicability of using a UV light source with the DBD reactor.
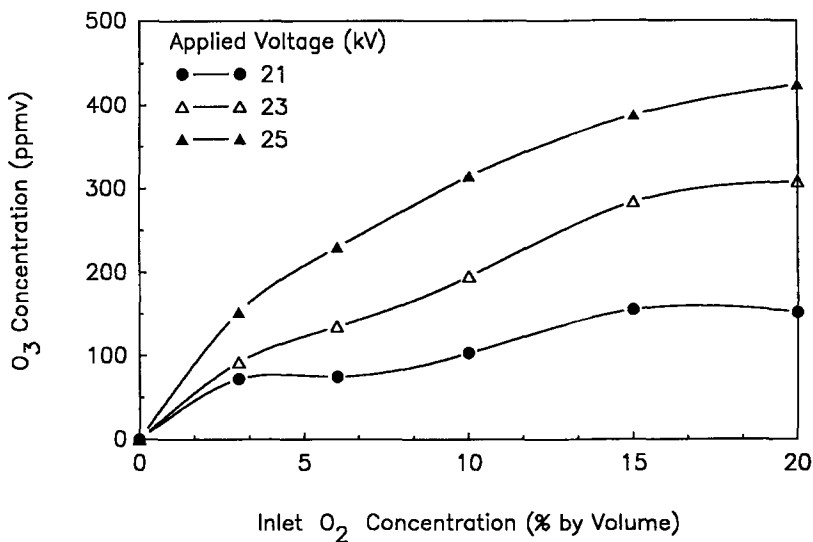
FIG. 3. Dependence of Resulting O_3 Concentration on Inlet O_2 Concentration and Voltage Applied to DBD Reactor

The dependence of $[O_3]$ on inlet $[O_2]$ at three voltages applied to the DBD reactor is presented in Fig. 3 using N_2 as the carrier gas. The gas stream was dry $\{[H_2O_{(g)}] < 3$ parts per million by volume (ppmv)$\}$, at room temperature ($\approx 25°C$), and at a flow rate of 2.7 L/min at standard conditions of $0°C$ and 101 kPa (1 atm). For these tests, the O_3 detector was located immediately downstream of the DBD reactor. At 25 kV, $[O_3]$ increases up to 425 ppmv with inlet $[O_2]$ increasing from 0% to 20% by volume. $[O_3]$ produced in the DBD reactor is less than the $[O_3]$ typically achieved by O_3 generators (up to a few thousand ppmv) because the reactor was not optimized for O_3 generation. In particular, the effective electrode gap was larger than that commonly used for ozonizers (≈ 20 mm compared to a few mm). The configuration of the DBD reactor was based on a low differential pressure across the reactor as needed for large-scale flue gas cleanup applications, <10 cm $H_2O_{(l)}$. The amount of O_3 generated at elevated temperatures is expected to be less than at the conditions tested, but was not characterized due to limitations of operating the O_3 detector at elevated temperatures with gas streams containing SO_2, NO, and NO_x. However, the DBD reactor can generate at least 400 ppmv of O_3 at the previously described conditions, which is significant relative to typical values of $[SO_2]$ and [NO] in flue gas streams of about 1,000 ppmv and 250 ppmv, respectively.

Measurements of η_{SO_2} achieved by DBD and CPP were initially completed at 130°C with gas streams containing 1,000 ppmv SO_2, 6% by volume O_2, 12% by volume CO_2, and N_2 as the carrier gas. $[H_2O_{(g)}]$ of the gas stream ranged from 0 ($[H_2O_{(g)}] < 3$ ppmv) to 9% by volume (Fig. 4). These concentrations are similar to the compositions of gases generated by fossil-fuel combustors. As shown, UV radiation does not significantly enhance η_{SO_2} achieved by DBDs when the gas stream is dry. With 9% by volume $H_2O_{(g)}$ and an applied voltage of 25 kV, η_{SO_2} achieved by CPP is about 18% larger
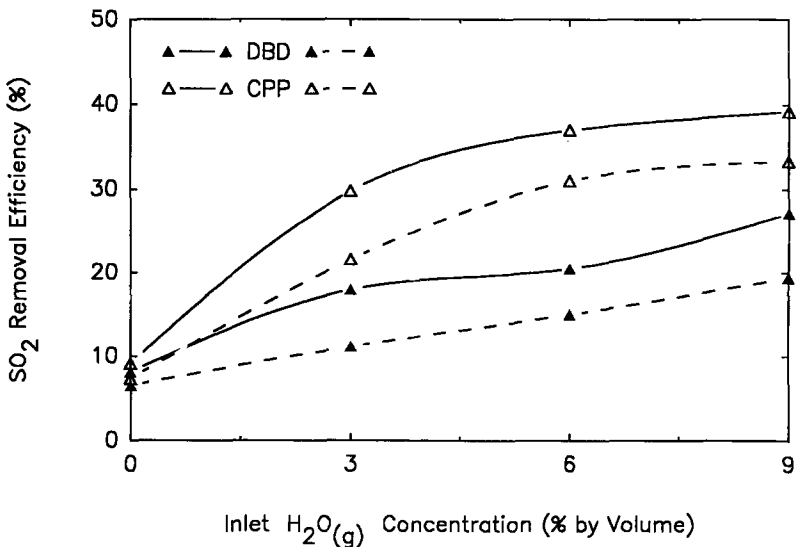
FIG. 4. Dependence of η_{SO_2} on inlet $[H_2O_{(g)}]$ at 130°C; Tests are Performed with DBDs and CPP (Dashed and Solid Lines Represent Applied Voltages of 23 kV and 25 kV, Respectively)

than that achieved by DBDs. η_{SO_2} achieved by CPP approached 39% for a gas stream containing 9% by volume $H_2O_{(g)}$. However, η_{SO_2} ranged from 1% to 12% with $[H_2O_{(g)}]$ ranging from 0% to 9% by volume, respectively, when UV radiation was used without the application of DBDs (Chang 1992). These results indicate that increased values of η_{SO_2} by CPP are not entirely attributed to the more efficient generation of OH radicals resulting from the photolysis of O_3 by UV radiation, as described by reactions (3) and (4). Direct photolysis of SO_2 to SO and direct photolysis of $H_2O_{(g)}$ to form OH radicals may also contribute to the enhancement of η_{SO_2} achieved by CPP when compared to using DBDs only (Welge 1974; Sethi 1971; Finlayson-Pitts and Pitts 1986).

It is also important to consider the effectiveness of CPP to remove SO_2 from gas streams that have different levels of $[CO_2]$. Experimental results have shown that η_{SO_2} increases with decreasing $[CO_2]$ such that η_{SO_2} increases by 107% (η_{SO_2} increasing from 14% to 29%) with $[CO_2]$ decreasing from 12% to 0% by volume (Chang et al. 1992b). Such observation is important for combustors that operate with flue gases that contain $[CO_2]$ <12% by volume.

To evaluate the influence of UV radiation on η_{NO}, DBD-generated plasmas were also illuminated by UV radiation when NO was in the gas stream. Experiments were completed at 130°C with gas streams containing 250 ppmv NO, 6% by volume O_2, 12% by volume CO_2, and N_2 as the carrier gas (Fig. 5). $[H_2O_{(g)}]$ ranged from 0% to 9% by volume. Experimental results show that instead of enhancing η_{NO}, UV radiation with DBDs decrease η_{NO} when compared to using DBDs only. For instance, with 9% by volume $H_2O_{(g)}$ in the gas stream and an applied voltage of 25 kV, η_{NO} decreases from 88% with DBDs only to 83% with CPP. Decrease in η_{NO} with CPP is attributed to photolysis of $HNO_{3(g)}$ to NO_2, NO_2 to NO, and HNO_2—an intermediate
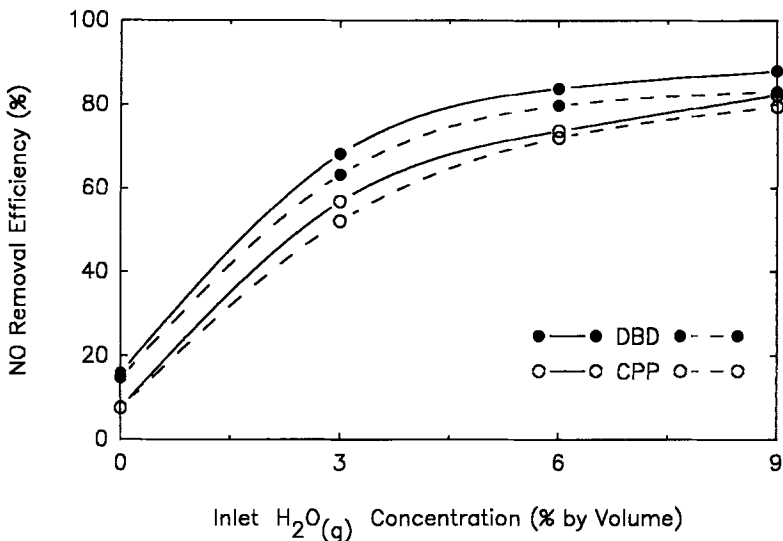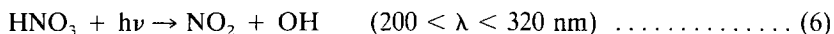
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FIG. 5. Dependence of η_{NO} on Inlet [$H_2O_{(g)}$] at 130°C (Dashed and Solid Lines Represent Applied Voltages of 23 kV and 25 kV, Respectively)

species formed by reaction of NO with OH (Chang 1992)—to NO as shown in the following reactions (Johnston and Graham 1973; Harvey et al. 1977; Baulch et al. 1982), respectively.

$$HNO_3 + h\nu \rightarrow NO_2 + OH \quad (200 < \lambda < 320 \text{ nm}) \dots\dots\dots\dots\dots (6)$$

$$NO_2 + h\nu \rightarrow NO + O \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots (7)$$

$$HNO_2 + h\nu \rightarrow NO + OH \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots (8)$$

The effect of UV radiation on [NO] was also determined without the generation of DBD plasma. While [NO_x] of the gas stream does not change appreciably with UV radiation, the ratio of [NO] to [NO_x] increases by a few percent with [$H_2O_{(g)}$] increasing from 0% to 9% by volume (Chang 1992). This result indicates that UV radiation photolyzes some of the NO_2 existing in the gas stream to form NO as shown in reaction (7) and possibly by the chemical reduction of NO_2 by O to form NO and O_2 (Chang 1992).

Experimental tests were also completed to evaluate the effect of using UV radiation and DBDs on η_{SO_2} and η_{NO} when SO_2 and NO coexist in the gas stream (Fig. 6). Experiments were completed at 130°C with gas streams containing 0% to 9% by volume $H_2O_{(g)}$, 1,000 ppmv SO_2, 250 ppmv NO, 6% by volume O_2, and 12% by volume CO_2 with N_2 as the carrier gas. The applied voltage was 25 kV. In the dry gas stream, CPP does not enhance η_{SO_2} beyond η_{SO_2} achieved by DBDs only, but decreases η_{NO} achievable by DBDs only. As [$H_2O_{(g)}$] increases, the enhancement of η_{SO_2} by CPP also increases. For a gas stream with 9% by volume $H_2O_{(g)}$, η_{SO_2} achieved by CPP is 29%, which is an increase of 26% compared to η_{SO_2} of 23% achieved by DBDs only. However, η_{NO} achieved by CPP is 79%, which is 9% less than η_{NO} of 87% achieved by DBDs only.

Values for η_{NO_x} and η_{NO} were very similar except the values for η_{NO_x} were 10% to 15% lower than for η_{NO}. Difference in values suggests that
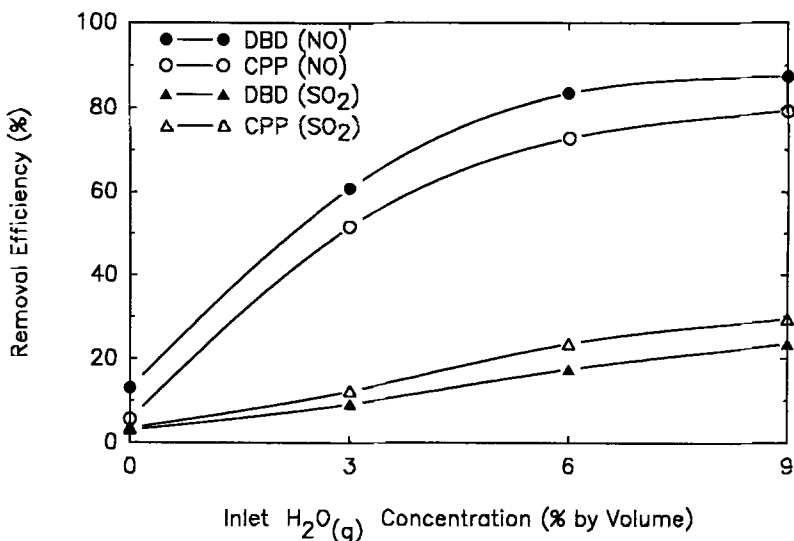
FIG. 6. Dependence of Simultaneous η_{SO_2} and η_{NO} on Inlet [$H_2O_{(g)}$] at 130°C; η_{SO_2} and η_{NO} Are Achieved Initially with DBDs and Then with CPP (Applied Voltage is 25 kV)

about 5% to 10% of the NO was converted to NO_2 instead of HNO_3 (Chang et al. 1992b).

Practical considerations of CPP include gas residence time and energy requirements. The gas residence time in the plasma was about 5 sec depending on the temperature of the gas stream. Such gas residence time is favorable compared to gas residence times of about 10 sec for conventional wet scrubbers and electrostatic precipitators. Energy consumption for CPP has been estimated at 10's of mJ per cm^3 (Chang et al. 1991). Such energy requirement is equivalent to 7% of the gross electrical generating capacity of a 500 MW coal-fired combustor. The energy requirement for a conventional SO_2 wet scrubber is about 3% of the gross electrical generating capacity for the same facility.

Although η_{SO_2} is below 70% as needed for new source performance standards for coal combustion ("Title" 1992), there has not been a concerted effort to optimize the reactor design or to increase the reactor's gas residence time. Such efforts may prove to increase η_{SO_2} and η_{NO} significantly without increasing energy requirements.

SUMMARY AND CONCLUSIONS

The effectiveness of combined plasma photolysis (CPP) to simultaneously remove SO_2 and NO from simulated flue gas streams was evaluated experimentally with a laboratory-scale reactor. Results obtained at 130°C show that CPP can enhance the removal efficiency of SO_2 (with no NO_x in the gas stream) to 39% when compared to a SO_2 removal efficiency of SO_2 of 33% with dielectric barrier discharges (DBDs) only. Removal efficiency of NO (with no SO_2 in the gas stream) achieved with CPP is 83% compared to an NO removal efficiency of 88% achieved with DBDs only. Simultaneous

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removal efficiencies for SO_2 and NO at inlet concentrations of 1,000 ppmv and 250 ppmv are as high as 29% and 79%, respectively.

Further research to optimize the geometry of the reactor and gas residence time is important to make the system more competitive with energy requirements for exiting wet scrubbing systems. More detailed measurements and analysis of the gas-phase intermediates and reaction endproducts would be useful. For example, measurement of the gas-phase species with a gas chromatograph–mass spectrometer would provide better insight into the dominant pathways taken to oxidize SO_2 and NO. Lastly, the influence of flyash in removing SO_2 and NO from gas streams may be significant and should be characterized.

APPENDIX.   REFERENCES

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