Evaluation of NO_x Removal by Corona Induced Non-Thermal Plasmas

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This paper discusses the characteristics of streamer corona and NO removal in air with pulsed and simplified DC/AC power supplies. In simplified gaseous mixtures of N₂+NO and N₂+O₂+NO and at room temperature, the energy efficiencies for NO removal are the same order of the magnitude with pulsed and simplified DC/AC power supplies. Near 100% of NO could be removed from air with a corona specific energy density of 2Wh/Nm³ for lower NO concentrations (NO<25ppm). DC/AC energized streamer corona may be one of the most cost effective and commercial available techniques for producing non-thermal plasma with larger electrodes gap distance (>5cm). Moreover, a detailed comparison between various corona induced non-thermal plasma techniques is presented.

Keywords: Streamer Corona, Electron Energy, Chemical Kinetics, Power Supply, Non-Thermal Plasma, NO_x Removal

1. Introduction

Non-thermal plasmas are usually dry processes and can be produced by various kinds of gaseous discharges, such as pulsed streamer corona, microwave discharge, gliding arc, glow, dielectric barrier and surface discharge. For larger gap (>5cm) electrode arrangements, it seems that positive pulsed streamer corona is very suitable for producing non-thermal plasma under high pressure. As one of the post-combustion cleaning techniques to remove NO_x and SO₂ from exhaust gases, corona induced non-thermal plasmas have demonstrated its technical and economical feasibility in contrast to commercially available techniques⁽¹⁾. According to the pilot plant test, it was estimated that for DeNO_x and DeSO₂ from coal fired flue gases by positive pulsed streamer corona the voltage pulse generator would cost about 80% of the investments although the concrete pulsed power techniques are not available yet. Moreover, the operation cost is mainly due to the total energy consumption^(1,2).

Most of recent experimental investigations are focused on increasing the energy efficiency, optimizing energization⁽³⁻⁶⁾ and enhancing heterogeneous reactions⁽⁵⁻⁸⁾. The energy cost for NO_x removal could be reduced from about 50-200eV/NO for the pilot plant tests to about 20eV/NO for laboratory tests. However, very few techniques have been reported for reducing the cost of voltage pulse generator and/or producing pulsed streamer corona⁽⁹⁻¹²⁾.

For promoting industrial applications of DeNO_x & DeSO₂ by pulsed streamer corona induced non-thermal plasma techniques, much more reliable pulsed power techniques are required for

producing streamer corona. Moreover, following several issues must be also considered: removal efficiencies, by-products composition, fabrication and lifetime of devices, cost of equipment and the total energy consumption. At the moment, the expensive method of producing streamer corona becomes the main block for industrial applications of flue gas cleaning.

Chang proposed three approaches for plasma treatments according to oxidation and reduction characteristics and treatment procedures⁽¹³⁾. Most of experimental investigations on NO_x, SO₂ removal are based on the oxidation processes to convert NO_x, SO₂ to their acids and/or salts with additives, such as NH₃. Only a few works were reported for NO_x reduction treatments, which are usually based on nitrogen plasma jets⁽¹⁴⁾ and plasma assisted NH₃ injection⁽¹⁵⁾. For oxygen-rich exhaust gases, plasma enhanced catalytic reduction of NO_x is investigated^(16,17). For E-beam processing, it was estimated that about 20% of removed NO is reduced to N₂ via NH₂ and N radicals induced reduction reactions, 10% to N₂O and about 70% firstly to NO₂ and then to NH₄NO₃, respectively⁽¹⁸⁾. Civitano estimated that for 15% of NO_x removal induced by radicals reaction with pulsed streamer corona treatment in coal fired flue gas, 70% of NO is converted to: NO₂ (50%), HNO₃ (10%) and N₂O (5%), respectively⁽²⁾.

As one of our continuous work on fundamental investigations on NO_x removal^(6,10-12), this work also reports our recent fundamental studies on characteristics of streamer corona production and NO removal with pulsed and simplified DC/AC power supplies. A detailed experimental investigation on oxidation and reduction processes during NO_x removal by corona induced non-thermal plasmas is reported in a separate paper⁽¹⁹⁾.

2. Characteristics of streamer corona and NO removal

Recent investigations on pulsed streamer corona are mainly based on advanced electrical and optical measurements^(4,6,20,21), and comprehensive streamer models^(22,23). Within a few hundreds of nano-seconds, ions energy transfer into reactor is very small in contrast to electron energy transfer⁽²⁴⁾, which leads almost all researchers to develop nano-seconds power generator for producing non-thermal plasmas. However, how much energy is transferred into reactor via ions current during pulsed streamer corona treatments is fully understood.

On the other hand, it has been well known that for centimeters gap point-to-plate electrodes in air, the morphology of corona discharge changes from onset streamer to Hermstein glow, pre-breakdown streamer and then to spark breakdown with increasing the positive DC applied voltage⁽²⁵⁾. With laser⁽²⁶⁾, X-ray⁽²⁷⁾ pulse excitation and changing gaseous compositions⁽²⁸⁾ or gas flow velocity⁽²⁹⁾, streamer corona can be also generated from a DC steady state glow discharge. For producing very large volume of streamer corona with a DC power supply, both glow discharge and streamer corona may be generated simultaneously as observed with a two different nozzles electrode⁽³⁰⁾ and a wire-to-plate reactor as shown in Fig.1, for which glow discharge induce negligible chemical reactions^(12,28). Generating uniform streamer corona with simplified power supplies has been one of our recent research objects.

Fig.1: Stationary photograph of positive DC corona in a wire-to-plate reactor. The diameter of high voltage electrode is 140 μ m.

2.1 Nano-seconds pulsed power supply

Several electric circuits have been proposed for producing voltage pulses of high amplitude, short duration and very short rise time. The most common circuit is based on the discharge of a capacitor in a low inductance circuit through a spark gap⁽⁶⁾. In order to show the basic characteristics of pulsed streamer corona under pulsed power supplies, Fig.2 shows a typical example of the time resolved SPS(0,0) emission of N₂ at 337.1nm and FNS(0,0) emission of N₂⁺ at 391.4 nm along the wire electrode in a wire-to-cylinder reactor⁽³¹⁾. The first peaks are associated to primary streamers and the second peaks are related to both secondary and newly produced primary streamers. The inbetween roughly corresponds to the moments that primary streamer arrives at the cylinder⁽⁶⁾. Pulsed streamer coronas in air and in flue gases show very similar structure and streamers propagate from the anode to the cathode as the primary and secondary streamers. With increasing the applied voltage, primary streamer may cross the gap, and then secondary streamer starts to develop when the primary streamer approaches the

Fig.2: Time resolved streamer light emission near the anode in air under various applied voltages V_p with a DC bias $V_{dc}=30$ kV in a wire-to-cylinder reactor.

cathode. In terms of the emission intensities, the derived electron energy of about 10eV for streamer heads^(20,31), which is much larger than the energy for secondary streamer. The streamer properties are also very similar to the self-sustained streamer corona under a DC power supply^(32,33). If the applied voltage is too small for streamer channel to bridge the gap, or the pulse duration is just long enough for primary streamer propagation, only primary streamer is observed. The produced streamer coronas always show such kind of characteristics for point-to-plate, wire-to-cylinder and wire-to-plate reactors. However, the energy transfer per each streamer depends on both power supply and reactor design⁽⁶⁾. In the reference⁽⁴⁾, the two phases of streamer corona are named as initial and resistive streamers, respectively. According to electron energy measurements^(20,31,32), one may argue that the main object of using pulsed power supply may be to suppress ion current instead of to increase the electron energy.

The reported energy cost for each NO molecule removal in literature ranges from 20eV/NO to 200eV/NO⁽⁶⁾. One of the reasons for such larger different energy costs is that the energy transfer ratios for the two phases of streamer corona are different

Fig.3: Typical experimental results in terms of the removed NO and the energy cost. (1) Civitano (1993) in coal fired flue gas; (2) Yan et.al (1998) in simulated flue gas; (3) Puchkarev et.al (1997) in diesel engine exhaust gas; (4) Veldhuizen et.al (1997) in methane fired flue gas; (5) Amirov et.al (1993) in methane fired flue gas; (6) Blom (1997) in air.

from one of the quoted tests to another. As an example, Fig.3 shows typical experimental results according to literature in terms of the removed NO and the energy cost^(2-5,34). For a given amount of NO removal, the energy consumption can be reduced by optimizing either the pulse width⁽²⁾, averaged corona current⁽³⁾ and the applied voltage level⁽¹¹⁾. Test conditions, such as gaseous compositions, gas temperature, treatment time, additives and catalyst can also significantly affect the efficiencies as shown in Fig.3^(5,6,35-37). The most energy efficient removal processes are usually achieved near the dew point, under high H₂O and O₂ concentration and with additives. For pulsed power supplies, the highest removal efficiency can be achieved when pulse duration equals to the time required by the primary streamer to cross the gap^(5,6). Based on streamer characteristics, a general principle for designing power supplies and plasma reactors was also proposed⁽⁶⁾. For an industrial size reactor, depending on the gap distance and applied voltage level, the pulse width may last a few tens of nano-seconds. By optimizing power supplies and treatment conditions, the energy costs in order of 20eV/NO for NO_x removal in flue gases could be achieved.

2.2 DC/AC power supply

For producing uniform streamer corona with simplified commercial available power supplies, a DC superimposed high frequency AC power supply (10kHz-60kHz) has been developed for NO_x removal^(10,12). According to electrical and optical measurements, the streamer corona produced with DC/AC power supplies possess very similar structure as pulsed streamer corona produced by using pulsed power supplies but streamers occur with lower energy density per pulse and very high repetition rates. Energy and charge transfers per each streamer are similar to the pulsed streamer corona with a peak current per unit length of wire of about 100A/m^(6,10,38). By increasing peak current per unit length of wire with pulsed power supplies, more than ten times

energy and charge transfers per each streamer can be also realized in contrast to DC/AC energized streamer corona. For larger electrodes gap distance, streamer channel energized by a pulsed power supply could extend to about 1 cm in diameter⁽⁴⁾, which may be much wider than the streamer channel energized with a DC/AC power supply. With a pulsed power supply almost all streamers are produced simultaneously⁽²⁰⁾, while streamers energized with a DC/AC power supply are randomly produced along the high voltage electrode⁽¹⁰⁾. A detailed comparison between the two methods of producing streamer coronas is summarized in Table 1.

Unlike using a DC power supply, streamer corona can be always produced provided the applied voltage covers the region of onset streamer with a DC/AC power supply. As a result, once streamer is generated, the Hermstein glow does not appear. Corona discharge also becomes less sensitive to electrodes misarrangements⁽³⁰⁾. Moreover, our recent experiments show that with the voltage change rate of 0.2kV/μs, DC glow may be transferred to streamer corona with a DC/AC power supply provided the AC peak voltage is larger than 1.0kV⁽¹²⁾.

Fig.4 shows the relationship between the total averaged corona current and ion current in terms of the averaged streamer current under a DC power supply, where an eight nozzles electrode is used. The averaged streamer current is calculated by multiplying the averaged streamer repetition rate with the averaged charge transfer of the streamer pulse⁽³⁸⁾. The ion current is defined as the difference between the total corona current and the averaged streamer current. Detailed experimental set-up and measurements system were reported in previous papers^(10,38). With increasing the applied voltage, the ion current and streamer current increase. The ion current is about 20% of the total current. With DC/AC power supplies, the ratio of ion current to the total current should be smaller in contrast to only using DC power supply⁽¹²⁾.

For evaluating chemical efficiency of DC/AC energized streamer corona, NO_x removal and NO to NO₂ conversion

Fig.4: Total corona and ion current in terms of streamer current under DC power supply and following conditions: [NO]=176ppm, [NO_x]=190ppm, [NH₃]=420ppm, [O₂]=19.5%, [CO₂]=0.7%, [N₂]=79.8%, F=5.7L/min; --- : zero ions current.

Fig.5a: Schematic of reactor and the saw-pipe electrode

Fig.5b: Schematic of the saw-pipe electrode

experiments have been extensively carried out in various simplified gaseous mixtures^(10,19) with nozzles and saw-pipe high voltage electrodes at atmospheric pressure and in room temperature. N and O radicals energy yields are also evaluated according to NO oxidation in simplified gaseous mixtures of N_2+NO and $N_2+O_2(\geq 3.6\%)+NO$, respectively⁽¹⁹⁾. As an example, Table 2 lists a comparison of N radicals production and NO removal in simplified N_2+NO gaseous mixtures with pulsed streamer corona^(37,39,40), dielectric barrier discharge⁽⁴⁰⁾ and DC/AC energized streamer corona⁽¹⁹⁾. The smaller energy cost of 70eV/N for N radicals production with pulsed streamer corona⁽³⁹⁾ may be due to the longer electrodes, smaller applied voltage, shorter pulse duration and higher initial NO concentration. For the experiments with pulsed streamer corona^(37,40) and dielectric barrier discharge⁽⁴⁰⁾, the energy costs for N radicals production are the same order with DC/AC energized streamer corona. It is estimated that within present test conditions the energy costs for producing each nitrogen radical in N_2+NO_x mixtures and oxygen radical in $N_2+O_2(\geq 3.6\%)+NO_x$ mixtures are about 170eV and 50eV, respectively⁽¹⁹⁾.

Fig.5 and Fig.6 show the schematic of the reactor, saw-pipe high voltage electrode and NO removal rate, respectively. Additional gases, such as NH_3 , Ar and CH_4 , can be also injected into reactor through the holes on the pipe via corona discharge region. The measurement system and the power supply are the same as the previous one^(10,12). A very uniform streamer corona can be generated inside the reactor with a maximum power density of 300W/m. Near 100% of NO could be removed from air with a corona specific energy density of 2 Wh/Nm³ for lower NO concentrations ($NO < 25ppm$) as shown in Fig.6. For 95% of NO removal under initial concentration of 46ppm, the energy yield is about 25g(NO)/kWh, which is the same order of the magnitude with pulsed power supplies⁽²⁸⁾. For larger NO_x concentration ($NO_x > 50ppm$) in air, the energy efficiency is significantly affected by the reverse reaction to convert NO_2 to NO via O radicals⁽¹⁹⁾.

Fig.6: NO removal rate in terms of the corona specific energy density in dry air under following conditions:
 $V_p = 3kV$, $f = 50kHz$, $V_{dc} = 0-30kV$

For NO_x removal from flue gases with additives and/or under larger humidity (near dew point), unlike in above simplified gaseous mixtures, both gas phase and heterogeneous phase reactions greatly contribute the removal efficiency. As a result, the most effective removal processes are observed in flue gases as shown in Fig.3. For DC/AC energized streamer corona techniques, there is no available experimental data to show its effectiveness in flue gases.

3. Conclusions and Discussions

Commercial available corona induced non-thermal plasma techniques are mainly based on dielectric barrier discharge for O_3 generation, which is driven by simplified AC power supplies⁽⁴¹⁾. Because of smaller electrodes gap, higher pressure drop across the reactor and the risk of insulator failure, these kinds of plasma reactors are less attractive for larger scale industrial NO_x , SO_2 treatment. The same remarks are also valued for packed-bed plasma reactor. With nano-seconds pulsed power supplies, NO_x could be very efficiently removed with an energy cost of about 20eV/NO. However, the development of the voltage pulse generator becomes the main block to industrial applications. With the DC/AC energization, the production of pulsed streamer corona can be significantly simplified without greatly reducing its chemical efficiency. Moreover, very energy efficient and cost-effective commercial power supplies are also available. Thus, a global equivalent energy efficiency with DC/AC energized streamer corona, which includes both energy conversion efficiency of power supply and plasma chemical efficiency, may be achieved in contrast to the energy efficiency with nano-seconds pulsed power supplies. However, future pilot plant investigations in real flue gases are required for evaluating its feasibility for flue gas cleaning. A general comparison of advantages and drawbacks of various corona induced non-thermal plasma techniques is listed in Table 3.

Table 1: Comparison of Pulsed Power and DC/AC Energized Streamer Corona

	Pulsed Power Energized Streamer Corona	DC/AC Energized Streamer Corona
pulse repetition rate	50-1500 pps	1 kHz-130 kHz
electron energy of streamer heads	~ 10 eV	~ 10 eV
peak current	≤ 600 A/m (per unit length of wire)	10 - 200 mA/ streamer
peak power	≤ 60 MW/m (per unit length of wire)	~ 3 kW/ streamer
EMC (di/dt)_max	≤ 30 A/ns/m (per unit length of wire)	~ 30 mA/ns/ streamer
average streamer velocity	5.0×10^5 - 3.5×10^6 m/s	~ 2.0×10^5 m/s
energy for primary streamer	≤ 3 J/m/pulse or 3 mJ/streamer	~ 0.3 mJ/ streamer
streamer diameter	200 μ m - 1 cm	?
ions current/total current	?	~ 20%
methods to control secondary streamer and spark breakdown	- using shorter voltage pulse - limiting applied voltage level	limiting applied voltage level
time delay between streamers	~ 3 ns (for a 50 mm length wire)	random (200 ns-1 ms for 100mm length wire)

Table 2: Comparison of energy cost for NO removal and N₂ production in N₂+NO mixtures by corona induced non-thermal plasmas

NO (ppm)	Temperature (°C)	Residence Time (S)	Flow Rate (L/min)	Length (mm)	Energy Density (Wh/Nm ³)	Energy Cost (eV/NO)	Energy Cost (eV/N)	Corona Mode	Ref
2000	100	18.8	4	1500	20.8 - 41.6	70	70	P.S.	39
100	100 - 300	0.78	65	300	≤ 27.8	240	240	P.S.	40
100	100 - 300	0.17	65	300	≤ 27.8	240	240	D.B.	40
1200	90 - 100	1.5	0.1	40		200	200	P.S.	37
440	20 - 25	5 - 10	4 - 20	100	≤ 5	85	170	D.S.	19

P.S.: Pulsed Streamer Corona; D.B.: Dielectric Barrier Discharge; D.S.: DC/AC Streamer Corona

Table 3: Comparison of various kinds of corona induced non-thermal plasmas

	Pulsed Power Induced Streamer corona	Dielectric Barrier Discharge	Packed-bed Corona Discharge	DC Streamer Corona Discharge	DC/AC Energized Streamer Corona Discharge
Advantages	- simple reactor - larger electrodes gap - small pressure drop - limited ions current - uniform plasma distribution	- simple and commercial available power supply - spark self quenching - less significant EMC noise	- simple and commercial available power supply - less significant EMC noise	- simple reactor - larger electrodes gap - small pressure drop - simple & commercial available power supply - less significant EMC noise	- simple reactor - larger electrodes gap - small pressure drop - simple and commercial available power supply - uniform plasma distribution - less significant EMC noise
Drawbacks	- expensive and no commercial available power supply - Significant EMC noise emission	- expensive reactor - high pressure drop - insulator failure	- expensive reactor - high pressure drop - short life time of pellets	- non- uniform plasma distribution (Glow & Streamer) - larger ions current	- larger ions current - need pilot plant demonstration

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