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NO Removal Characteristics of a Corona Radical Shower System Under DC and AC/DC Superimposed Operations

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Abstract—In this paper, the effects of the applied voltage modes on the positive corona discharge morphology and NO removal characteristics from air stream are experimentally investigated. By using a dc superimposed high frequency ac power supply (10–60 kHz), a uniform streamer corona can be generated, which is also less sensitive to electrode mis-arrangements. Hermstein glow can be transferred to streamer corona if the peak-to-peak voltage is larger than 1.0 kV at the voltage change rate of 0.2 kV/µs. A significant amount of NO removal is observed under streamer corona. For the Hermstein glow, the removal is negligible. Moreover, the basic principle for designing ac/dc energized streamer corona is also presented in this paper.

Index Terms—AC/DC superimposed operations, glow corona, NO removal, streamer corona.

I. INTRODUCTION

WITHIN the last 15 years, various kinds of corona-induced nonthermal plasmas have been investigated in order to remove gaseous pollutants, such as SO₂, NO₂, Hg, H₂S, and VOCs, from exhaust gases [1], [2]. These nonthermal plasmas are usually dry processes and can be produced by different kinds of gaseous discharges, such as pulsed streamer corona, microwave discharge, gliding arc, glow, dielectric barrier and surface discharge. Non-thermal plasmas can be also produced with wet electrode [3] or on the surface of catalyst [4]. Different kinds of additives, such as NH₃, H₂O₂, hydrocarbon (HC), N₂H₄ and natural gas, are injected into the plasma reactors for improving the energy efficiencies and controlling final by-products. Up to now, more than 50 kinds of pollution emission control with corona plasma techniques have been investigated [5].

For larger gap (≥5 cm) electrode arrangements, pulsed streamer corona is often used for producing nonthermal plasma under high-pressure. According to the pilot plant test at ENEL, Italy, it was estimated that for DeNOₓ and DeSO₂ from coal fired flue gases by positive pulsed streamer corona the voltage pulse generator would cost about 80% of the total investments although the concrete pulsed power techniques are not available yet. Moreover, the operation cost is mainly due to the total energy consumption [6]. However, very few work are available for discussing the requirements of pulsed power supplies and the design of plasma reactor for inducing chemical reactions [7]. For promoting industrial applications of DeNOₓ and DeSO₂ by pulsed streamer corona induced nonthermal plasma techniques, much more reliable pulsed power techniques are required for producing streamer corona [8]. Industrial demonstrations also become very important for evaluating technical and economical feasibility of the techniques.

On the other hand, it has been well known that for centimeters gap point-to-plate electrodes in air, the morphology of positive corona discharge changes from on-set streamer to Hermstein glow, prebreakdown streamer and then to spark breakdown by increasing the applied voltage [9], [10]. Using laser [11] and/or X-ray [12] pulsed excitation, streamer corona could be generated from a steady-state glow discharge. Discharge patterns can be also controlled by gaseous flow velocity and the injection gaseous compositions from hollow-type electrodes [13]–[15].

In order to limit NH₃ slip and improve the energy efficiency for NOₓ removal with dc streamer corona, a corona radical shower system was proposed by injecting additional gases of NH₃, N₂, O₂, CO₂, Ar and CH₄ from nozzles into electrode plasma [16], [17], where positive streamer propagates from the nozzles to the plate with a velocity in order of 2 × 10³ m/s [18]. SO₂, HCl and H₂S can be also effectively removed with such kind of system [19]. As one of our fundamental studies on characteristics of NOₓ removal by corona plasma technique [20], this work reports the characteristics of corona modes and NO removal with a corona radical shower system under dc and dc superimposed high frequency ac power supplies. The corresponding investigation on the oxidation and reduction processes during NOₓ removal by the corona plasma technique was reported elsewhere [21].

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II. EXPERIMENTAL SETUP

The schematic diagram of a corona radical shower system is shown in Fig. 1(a), where a pipe with multinozzles as shown in Fig. 1(b) is used as the active electrode and the plate electrode is used as the grounded electrode. The nozzle in perpendicular to the plate is connected to a 4-mm pipe, which is placed at the center of the reactor (125 × 100 × 500 mm³). The length, inner, and outer diameters of the nozzle are 5.0, 1.0, and 1.5 mm, respectively. Thus, the gap distance between the nozzle electrode and the grounded plate is 43 mm. The ac power generator is coupled to a positive dc power source with a 700-pF coupling capacitor [20]. The ac peak-to-peak voltage \( V_{pp} \) and the frequency \( f \) range from 0.5 kV (60 kHz) to 10 kV (10 kHz), respectively. For limiting full spark breakdown discharge, a 2.2-MΩ resistor is used between the dc power source and the reactor.

Streamer corona current and the applied high-voltage waveforms on the reactor are measured with Pearson current transformer (Model 150) and Iwatsu Voltage Divider (HV-130), respectively. The transformer and higher voltage divider are connected to the reactor with very short leads for very fast response measurements. Light emission between the electrodes during streamer propagation is observed with a quartz fiber of 300 μm in diameter and a PMT (Hamamatsu Type R5113), where the fiber is contained in a thin tube in order to focus the light rays from an expected region. These time-resolved signals are recorded with digital oscilloscope of LeCroy 9362 (10 GS/s, 1.5 GHz) and/or HP 54522A (2 GS/s, 500 MHz).

Time averaged corona current is measured with a current meter in series with a 1 kΩ resistor and in parallel with a 10 μF capacitor. The averaged current is used to evaluate the time averaged corona power and to calculate the corona specific energy density, which is defined as the ratio of the average corona power to the total gas flow rate. NO and NO (NO + NO) concentrations are measured with a nondispersive infrared analyzer (Horiba ENDA 1400).

CO₂ and/or N₂ + (20%)O₂ could be injected into the reactor through the nozzle electrode. Experiments are carried out with an eight-nozzle electrode under one atmospheric pressure and in room temperature for gaseous mixtures of N₂ + O₂ + NOₓ. Initial NOₓ concentration is between 50 and 100 ppm. Within the present work, the total gas flow rate \( Q_g \) and the additional
Fig. 2. Current and voltage characteristics under dc and ac/dc operations under following conditions: ac power supply: \( V_{pp} = 3 \text{ kV} \), \( f = 50 \text{ kHz} \), \([\text{O}_2]\) = 17\%, \([\text{N}_2]\) = 83\%, \([\text{NO}]\) = 50 ppm, \([\text{NO}_2]\) = 54 ppm. Addition gas flow rate \( Q_a = 1.0 \text{ L/min} \), Total gas flow rate \( Q_t = 3.5 \text{ L/min} \).

gas flow rate \( Q_a \) through the nozzle electrode are less than 10.0 and 3.0 L/min, respectively.

Glow and streamer coronas can be distinguished by means of electrical and optical measurements. For streamer corona discharge, both the current and voltage waveforms show pulsed characteristics and light emission can be observed inter the electrodes gap due to the streamer propagation. However, for Hermstein glow, the current and voltage waveforms show very even characteristics, and the corresponding light emission can be only observed near the tip of the nozzle.

III. RESULTS AND DISCUSSION

A. Corona Modes and Chemical Reactivity

Fig. 2 shows typical time averaged current–voltage characteristics under dc and ac/dc operating modes. Due to slight differences of nozzles geometry and gaseous compounds distribution near the nozzles, the dc onset of streamer corona voltage \( V_{cl} \) and the Hermstein glow voltage \( V_{gl} \) [15] are different from one nozzle to another. With increasing the applied voltage, the time averaged current–voltage curve shows a hysteresis under the dc power supply. According to the light emission, voltage and current waveforms, the two separate parts of the curve as indicated in Fig. 2 correspond to streamer corona and Hermstein glow, respectively. With increasing the applied voltage, onset streamer is transferred to the Hermstein glow at the largest transition voltage \( V_{cl} \). While, with decreasing the applied voltage, the Hermstein glow is transferred to streamer corona at the smallest transition voltage \( V_{gl} \). With the ac/dc superimposed operation, streamer corona can be always generated provided the applied voltage covers the regions of onset streamer. As a result, once streamer is generated, the Hermstein glow does not appear. Corona discharge also becomes less sensitive to electrode misarrangements [22].

Fig. 3. Dependence of corona mode and NO concentration under the same conditions as in Fig. 2.

[20]. The hysteresis may be due to the interinfluences between streamers from different nozzles and the thermal effects of the nozzles electrode.

The corresponding NO concentration is shown in Fig. 3 in terms of the corona specific energy density. NO concentration shows very different dependencies on the corona specific energy density because of the changes of corona modes. For glow corona mode, the removed NO is negligible. While, a significant amount of NO removal is observed under streamer corona mode. The same phenomenon of corona modes and NO removal in dry air was also observed when changing corona modes by CO2 injection [15]. Experiments in N2 + O2 + CO2 + NOx + NH3 gaseous mixtures under a dc power supply also show that higher NOx removal rate can be only achieved when a higher frequency dc self-sustained streamer corona is generated [18].

N and O radicals energy yields by ac/dc energized streamer corona are also evaluated according to the NO to NO2 conversion in simplified gaseous mixtures [21]. It is estimated that within present test conditions the energy costs for producing each nitrogen radical in N2 + NOx mixtures and oxygen radical in N2 + O2(>3.6%) + NOx mixtures are about 170 and 50 eV, respectively. The energy costs for N radical production are the same order as with pulsed streamer corona [23], [24] and dielectric barrier discharge [25].

Fig. 4 shows effects of the ac peak-to-peak voltage \( V_{pp} \) on the corona discharge modes, time averaged corona current and NO concentration under a dc bias voltage of 24.2 kV and the ac frequency of 50 kHz. For 1.0, 2.0 and 3.0 kV of peak-to-peak voltage \( V_{pp} \), the corresponding total voltages on reactor are 23.7–24.7 kV, 23.2–25.2 kV, and 22.7–25.7 kV, respectively. Within the tests, the Hermstein glow can be transferred to streamer corona by superimposing the ac voltage on the dc bias. However, with refer to the Fig. 2, one may see that in these tests the applied voltage does not cover the region of onset streamer, where the largest transition voltage \( V_{gl} \) is about 21 kV. When glow is transferred to streamer corona, the averaged current is increased from about 85 \( \mu \text{A} \) to 135 \( \mu \text{A} \), and the removed NO is increased from less than 2 ppm to about 40 ppm.
Fig. 4. Dependence of corona mode and NO concentration on the power supply under following conditions: ac power supply: \( f = 50 \) kHz, Gaseous compositions: \([\text{O}_2] = 17\%\), \([\text{N}_2]\) = 83\%, \([\text{NO}] = 50 \) ppm, \([\text{NO}_2] = 54 \) ppm, \(Q_e = 1 \) L/min, \(Q_i = 3.5 \) L/min. Test no. 1: \( V_{dc} = 24.2 \) kV, \( V_{pp} = 0 \) kV, corona mode—glow. Test no. 2: \( V_{dc} = 24.2 \) kV, \( V_{pp} = 3 \) kV, corona mode—streamer. Test no. 3: \( V_{dc} = 24.2 \) kV, \( V_{pp} = 2 \) kV, corona mode—streamer. Test no. 4: \( V_{dc} = 24.2 \) kV, \( V_{pp} = 1 \) kV, corona mode—streamer. Test no. 5: \( V_{dc} = 24.2 \) kV, \( V_{pp} = 0.5 \) kV, corona mode—glow.

Fig. 5. Dependence of NO concentration on the power supply with a eight nozzles electrode under the following conditions: \( V_{pp} = 26 \) kV, \( f = 60 \) kHz; \( V_{pp} = 3.8 \) kV, \( f = 40 \) kHz; \( V_{pp} = 7.2 \) kV, \( f = 20 \) kHz; \( V_{pp} = 10 \) kV, \( f = 10 \) kHz; \([\text{NO}] = 86 \) ppm, \([\text{NO}_2] = 90 \) ppm, \([\text{CO}_2] = 2.7\%\), \([\text{O}_2] = 14.8\%\), \([\text{H}_2\text{O}] = 1.0\%\), \([\text{N}_2] = 81.5\%\), \(Q_e = 1.1 \) L/min, \(Q_i = 4.0 \) L/min.

With injecting CO\(_2\) into the reactor through nozzles electrode, a uniform streamer corona could be also produced without producing Hermstein glow under a dc power supply [15]. In these cases, the energy efficiency of NO removal can be slightly improved with the ac/dc superimposed operation within present test conditions. Fig. 5 shows an example of the NO removal rate in terms of the corona specific energy density under dc and ac/dc power supplies. The improvement of NO removal under ac/dc energization is maybe due to limiting ion movement, which is also in agreement with the experiments presented in Fig. 3.

### B. Principles of AC/DC Corona Plasma Energization

According to numerical analysis on positive glow discharge, Morrow concluded that if the change rate of the applied voltage is larger than 1 kV/\(\mu\)s, glow would not appear with increasing the applied voltage [26]. Present experiments show that even with the change rate of 0.2 kV/\(\mu\)s of the ac voltage superimposed on a dc bias, the Hermstein glow can also be transferred to streamer corona if the ac peak-to-peak voltage is larger than 1.0 kV. With a dc superimposed pulsed power supply, streamer corona can be always generated provided the total peak voltage on the electrodes is larger than the inception voltage. However, the peak streamer current would be reduced if the dc bias becomes larger than the dc corona onset and a dc corona is generated [27]–[29]. For present dc superimposed ac power supply, the dc glow discharge does not show very significant effects on the streamer peak current, but it could greatly affect the streamer repetition rate after superimposing the ac power supply. In [26], the production of streamer corona from a steady-state Hermstein glow discharge was also reported by superimposing a 100-ns duration voltage pulse on a dc bias in a point-to-plate electrode arrangement. With regard to the capital cost of pulsed power generators, the ac/dc energized streamer corona may be one of the cost effective and commercial available techniques for producing streamer coronas. The transition from glow to streamer corona depends on the dc bias \((V_{dc})\), ac peak-to-peak voltage \((V_{pp})\), ac voltage waveforms, the frequency and gaseous compositions. Larger voltage change rate and ac peak-to-peak voltage are suitable for definitely transferring the glow discharge to streamer corona. Because the values of the specific voltages such as the streamer corona onset, the transition voltages from onset streamer to glow, from glow to pre-breakdown streamer, and the spark voltage depend on electrode geometry and the gaseous compositions, it seems difficult to produce larger volume uniform streamer corona under a given constant dc applied voltage. For the ac/dc power supplies, in principle, the voltage levels of \(V_{dc}\) and \(V_{pp}\) can be always adjusted to cover the regions of either onset or pre-breakdown streamers. As a result, a uniform streamer corona could be always produced.

### IV. Conclusion

An experimental investigation has been conducted to convert NO to NO\(_2\) in dry air with dc and superimposed ac/dc energized streamer coronas and the following conclusions are obtained.

1) During each cycle of ac/dc voltage, the applied voltage may cover all regions of onset streamer and Hermstein glow. As a result, streamer corona could be always generated without Hermstein glow. Streamer corona also becomes less sensitive to electrode mis-arrangements and/or variations of gaseous compositions.

2) In dc glow discharge region, glow can also be transferred to streamer corona provided the ac peaks voltage is larger than 1.0 kV at the voltage change rate of larger than 0.2 kV/\(\mu\)s.

3) ac/dc energized streamer corona may be one of the cost effective and commercial available techniques for producing corona plasmas with larger electrodes gap distance.
4) For Hermstein glow discharge, the induced NO conversion is negligible, while for streamer corona produced either by dc or ac/dc power supplies, a significant amount of NO conversion is observed.

REFERENCES


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