NO removal characteristics of a corona radical shower system under DC and AC/DC superimposed operations


Published in:
IEEE Transactions on Industry Applications

DOI:
10.1109/28.952527

Published: 01/01/2001

Citation for published version (APA):
NO Removal Characteristics of a Corona Radical Shower System Under DC and AC/DC Superimposed Operations

Keping Yan, Takashi Yamamoto, Seiji Kanazawa, Member, IEEE, Toshikazu Ohkubo, Member, IEEE, Yukiharu Nomoto, Member, IEEE, and Jen-Shih Chang, Senior Member, IEEE

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/ps. 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, NOₓ, 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 electrode into the reactor [16], [17], where positive streamer propagates from the nozzles to the plate with a velocity in order of 2 × 10² 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 using 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 plasma corona technique was reported elsewhere [21].
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 ($125 \times 100 \times 500$ mm$^3$). 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-M$\Omega$ 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 $\mu$m 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 k$\Omega$ resistor and in parallel with a 10 $\mu$F 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$_x$ (NO + NO$_2$) concentrations are measured with a nondispersive infrared analyzer (Horiba ENDA 1400).

CO$_2$ and/or N$_2$ + (20%)O$_2$ 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 N$_2$ + O$_2$ + NO$_x$. Initial NO$_x$ concentration is between 50 and 100 ppm. Within the present work, the total gas flow rate $Q_g$ and the additional...
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 \( V_s \) and the Hermstein glow voltage \( V_g \) [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 \( V_{g1} \). While, with decreasing the applied voltage, the Hermstein glow is transferred to streamer corona at the smallest transition voltage \( V_{g2} \). 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].

III. RESULTS AND DISCUSSION

A. Corona Modes and Chemical Reactivity

[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 CO\(_2\) injection [15]. Experiments in N\(_2\) + O\(_2\) + CO\(_2\) + NO\(_x\) + NH\(_3\) gaseous mixtures under a dc power supply also show that higher NO\(_x\) 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 NO\(_x\) conversion in simplified gaseous mixtures [21]. 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\(_2\) (>3.6%) + NO\(_x\) 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 \( V_{g1} \) is about 21 kV. When glow is transferred to streamer corona, the averaged current is increased from about 85 \( \mu \)A to 135 \( \mu \)A, and the removed NO is increased from less than 2 ppm to about 40 ppm.
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/µs, glow would not appear with increasing the applied voltage [26]. Present experiments show that even with the change rate of 0.2 kV/µs 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/µs.

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**


Keping Yan was born in China in 1962. He received the B.Sci. and M.Sci. degrees in applied physics from Beijing Institute of Technology, Beijing, China, in 1983 and 1986, respectively. He has been with Eindhoven University of Technology, Eindhoven, The Netherlands, since 1988, where his focus has been research and development of corona plasma techniques for pollution control. He has coauthored about 60 publications.

Takashi Yamamoto was born in Japan in 1974. He received the B.E. and M.E. degrees from Oita University, Oita, Japan, in 1997 and 1999, respectively. He currently is with Tokyo Electric Power Company, Ltd., Tokyo, Japan.

Seiji Kanazawa (M’91) was born in 1961. He received the B.E., M.E., and Ph.D. degrees from Kumamoto University, in 1985, 1987, and 1990, respectively. Since 1990, he has been with Oita University, Oita, Japan, where he is presently an Associate Professor in the Department of Electrical and Electronic Engineering. His research interests are in the areas of applied electrostatics, air pollution control, plasma technologies, plasma diagnosis and eco-material processing using lasers.

Dr. Kanazawa is a member of the Institute of Electrical Engineers of Japan, Institute of Electronics, Information, and Communication Engineers of Japan, Institute of Electrostatics, Japan, and Japan Society of Applied Physics.
Toshikazu Ohkubo (M’86) was born in Beppu, Japan, in 1948. He received the B.E., M.E., and D.E. degrees from Kyushu University, Fukuoka, Japan, in 1972, 1975, and 1986, respectively.

From 1975 to 1987, he was a Research Associate in the Department of Electrical Engineering, Oita University, Oita, Japan. From 1988 to 1989, he was a Visiting Research Fellow at McMaster University, Hamilton, ON, Canada. He became an Associate Professor in 1987 and a Professor in 1995 in the Department of Electrical and Electronics Engineering, Oita University. His research interests include NOx removal by corona discharge-induced plasmas, electrohydrodynamics, NO measurement by laser-induced fluorescence, and thin-film preparation by pulsed-laser deposition for fuel cell.

Dr. Ohkubo is a member of the Institute of Electrical Engineers of Japan, Institute of Electrostatics, Japan, and Japan Society of Applied Physics.

Yukiharu Nomoto (M’86) was born in Shinyo, China, in 1942. He received the B.E., M.E., and D.E. degrees from Kyushu University, Fukuoka, Japan, in 1965, 1967, and 1982, respectively.

In 1970, he became a Research Associate at Kyushu University. He joined Oita University, Oita, Japan, as an Associate Professor in 1973. He became a Professor in 1985. His research interests include low-temperature plasmas, plasma chemistry, electrostatic applications, and ozonizer.

Dr. Nomoto is a member of the Institute of Electrical Engineers of Japan, Institute of Electrostatics, Japan, and Japan Society of Applied Physics.

Jen-Shih Chang (M’90–SM’96) received the B.Eng. and M.Eng. degrees from Musashi Institute of Technology, Japan, and the Ph.D. degree from York University, Toronto, ON, Canada. During 1973–1974, he was a Researcher with the Centre de Recherches en Physique de l’Environnement, CNRS, France. From 1975 to 1979, he was a Project Scientist/Assistant Professor in the Department of Physics, York University. In 1979 and 1981, he was an Assistant and Associate Professor, respectively, in the Department of Engineering Physics, McMaster University, Hamilton, ON, Canada. During 1985–1995, he was a Visiting Professor at the University of Seville, Joseph Fourier University, The University Tokyo, Tokyo Denki University, and Musashi Institute of Technology. He is currently a Professor at McMaster University.