Application of micro plasma for ozone generation and environmental protection

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Abstract— Ozone generation of micro plasma is investigated which is occurred with a pair of electrodes covered with dielectric barrier. The discharge gap of micro plasma is in the order of micro meters, which is very small compared to other discharge methods. Since the discharge gap is very narrow, non-thermal plasma is occurred around 1 kV, which also produces high electric field between the electrodes. By using micro plasma, pollutant gas control is possible with a very small power source and system. In this paper, electrical characteristics of micro plasma, practical discharge power, and the effect of number of electrodes are experimentally investigated. Also, treatment of low concentration of hazardous gases, such as cigarette smoke, nitric oxide is carried out by using micro plasma. Byproducts are confirmed by FT-IR and Fragrance & Flavor Analyzer.

Keywords— micro plasma, ozone, NOx, cigarette smoke

I. INTRODUCTION

Ozone genratation at low discharge voltage and small power source is required for environmental protection for a practical use. Conventional nonthermal plasma is widely studied in science fields and also used for engineering fields, industry, and etc. Decomposition of air pollutants [1-6], gas synthesis and gas conversion [7-9] are examples of nonthermal plasma to enhance chemical reactions at atmospheric pressure. Although, these present nonthermal plasmas are generated by corona discharges and glow discharges, for which a high discharge voltage is necessary. By replacing this with dielectric barrier discharge, and using our micro plasma electrode with a discharge gap of micro meters, the discharge voltage is decreased to about 1 kV. Our nonthermal plasma is called micro plasma, since the plasma volume is very small. We have investigated micro plasma as a response to such needs [10,11].

We have concentrated on indoor air control, since indoor air control related concerns, such as sick-building syndrome, have been an environmental issue since 1970s world widely. There are many kinds of air pollution, and indoor air quality (IAQ) is a timely issue, since buildings and houses have become more airtight. For treating polluted air, low concentration of ozone plays an important role, which is generated by nonthermal plasma.

In this paper, the electrical characteristics of the micro plasma electrode are investigated. Also, generation of ozone and NOx characteristics and effect of number of electrodes is experimentally carried out for dry air.

Treatment of pollutant air such as cigarette smoke and nitric oxide is carried out by using micro plasma.

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A. About micro plasma

In order to generate micro plasma, we used a pair of perforated metal plates covered with dielectric materials as micro discharge electrodes. This pair of electrodes is faced together with a discharge gap of 10 um, which is maintained by placing a spacer between the electrodes [1, 2]. An alternating voltage of 25 kHz is applied to the electrodes, resulting intense streamers between the gaps. Since the discharge gap is small compared to that of other silent discharges [7], non-thermal plasma is occurred below a discharge voltage of 1 kV.

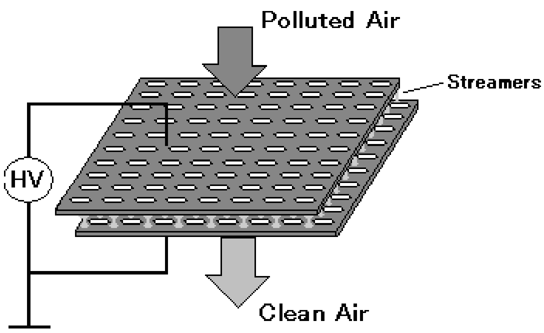


Fig. 1. Schematic image of polluted air control by using micro plasma electrode.

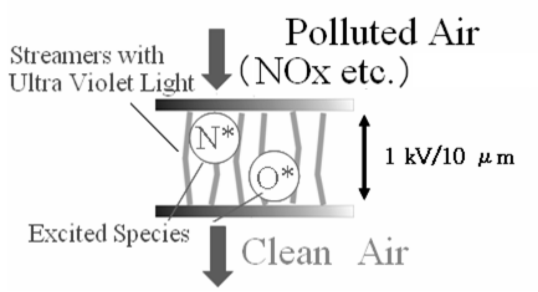


Fig. 2. A schematic image of the removal process of micro plasma between the electrodes.

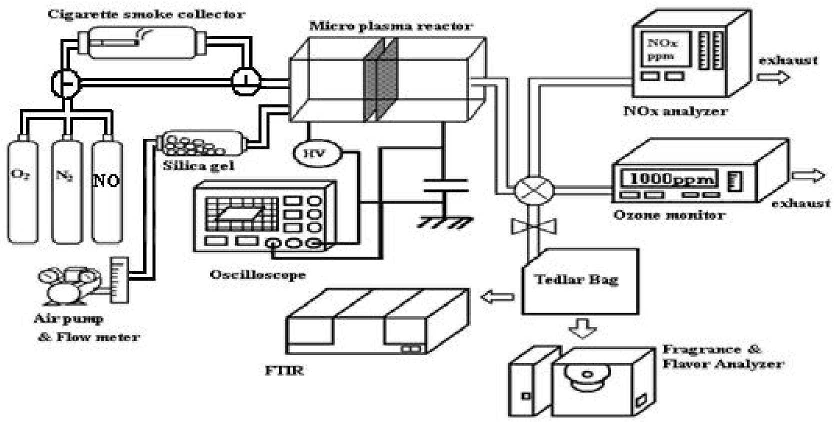


Fig. 3. Experimental setup for gas analysis and electrical measurement for micro plasma.

Fig. 1 shows a schematic image of the electrodes used in the experiments. It is a stainless steel covered with dielectric barrier materials both faces, and is 800 mm^2 in area. The aperture ratio is about 50%. The pressure loss between the electrodes is about 5 mmH_2O. The gas residence time between the electrodes is about 0.54ms at a gas flow of 10l/min. The gas is flowed through the holes and reacted with active electrons and other active species generated between the electrodes. The pressure loss is very small and the electrode could be applied to exhaust gas treatments with large gas flows.

When the discharge gap is set to less than 10 um, with a discharge voltage of less than 1 kV, high electrical field (10^7~10^8 V/m) can be easily obtained. High energy electrons and active species are generated in such micro discharges, which is activated by high electrical field.

II. Experimental setup

A. Analysis of treated gas

The experimental setup is shown in Fig. 3. Atmospheric pressure air is flowed to the micro plasma reactor through a silica gel packed-bed by an air pump and a flow meter, to investigate the ozone generation and NOx generation.

The gas mixture is also produced by using gas cylinders of N_2, NO, O_2 to compose accurate gas mixture. Concentrations of NOx and O_3 are measured by a chemiluminescence NOx analyzer (Shimadzu, NOA-7000) and an ultra violet adsorption type ozone monitor (Ebara, EG-2001B).

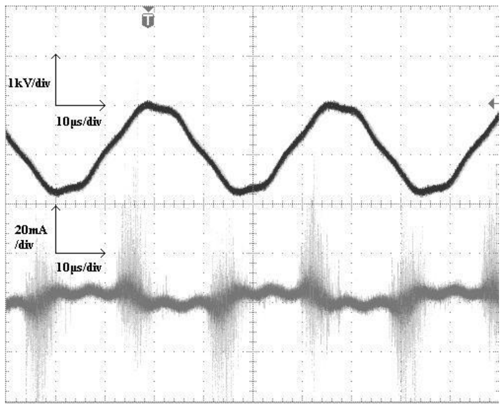
The treated gas was collected and analyzed by an FT-IR (Shimadzu Corporation, IRPrestige-21) and a Fragrance Flavor Analyzer (Shimadzu Corporation, FF-2A).

B. Electrical measurements and power source

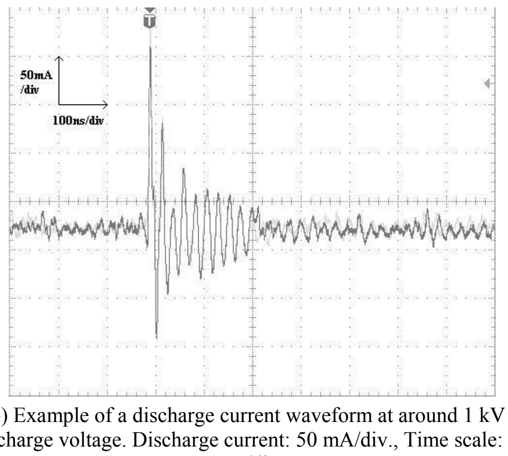
A high frequency transformer was used as the power supply of the micro plasma reactor, and its frequency is about 25 kHz.

The high voltage supplied to the electrode and its corresponding discharge currents are measured by a high voltage divider (Tektronix, P6015A), AC current probe (Tektronix, P6022) and a digital oscilloscope (Tektronix, TDS 3014). The discharge power is estimated by using the Lissajous method.

Fig. 4 shows a waveform example of a discharge voltage and discharge current. Spike currents, which



(a) Example of a discharge voltage and discharge current. Discharge voltage: 1 kV/div., Discharge current 20 mA/ div., Time scale: 10 us/div.



(b) Example of a discharge current waveform at around 1 kV of discharge voltage. Discharge current: 50 mA/div., Time scale: 100 ns/div.

Fig. 4. Waveforms of applied voltage and discharge current.

could be occurred by streamers convoluted on the current waveform, were confirmed in addition to the capacitive current at steepest slopes of the waveform. This is recognized as the discharge current shown in Fig. 4(b).

III. PRACTICAL DISCHARGE POWER OF MICRO PLASMA

Discharge power and power loss of the power supply is experimentally investigated to estimate the practical discharge power. Fig. 5 shows the relation between the discharge power and the discharge voltage for the micro plasma electrodes. The number of the electrode is increased up to 4 pairs to measure the differences of each discharge power of the electrode. The average of the difference between the discharge powers is the practical discharge power without the power loss. In addition, the loss of the power supply is calculated and plotted in Fig.5.

For example, the practical discharge power at 1080 V is about 6 W. Discharge power could be affected by streamer density, though it is difficult to observe the streamers occurring in the very narrow gap between the electrodes.

By using a thermograph camera, the temperature distribution is observed which could be suggesting the thermo effect of the streamers. The temperature distribution is affected by intense streamers of the plasma (See Fig. 6.). In this case, it can be seen that the streamers are occurring equally throughout the discharge gaps. Fig. 6 suggested that when the input power is low, streamer density could be low. Increase of surface temperature of the electrode could be suggesting that the streamer density is also high when the input power is high.

By comparing, input power of 10 W (Fig. 6(a)-(c)) and 13 W (Fig. 6(d)-(f)), the surface temperature of the electrode is increased with higher input power. Intense streamer, which starts at about 1000V, and dielectric loss could be causing the heat of the electrode. In addition, the heat of the electrode is passed to the gas stream, controlling the temperature rise of the plasma reactor.

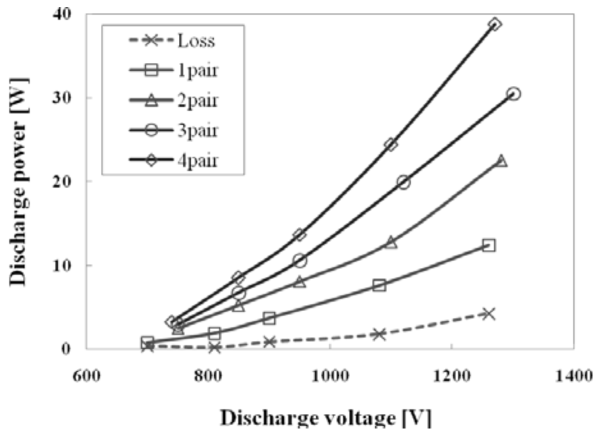


Fig. 5. Discharge power for various numbers of electrode pairs.

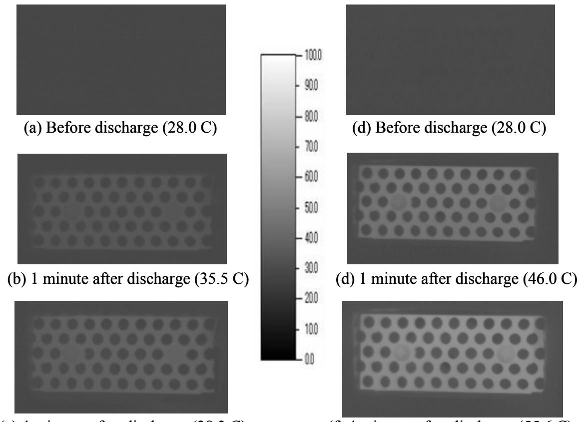


Fig. 6. Change of electrode temperature without flowing gas. Left column: (a)-(c). Input power of 10 W (Corresponding practical discharge power 2.5 W. Corresponding discharge voltage 800V). Right column: (d)-(f). Input power of 13 W (Corresponding practical discharge power 4 W. Corresponding discharge voltage 1000V).

IV. OZONE GENERATION BY MICRO PLASMA IN AIR

Measurements of ozone generation is considered to estimate the effect of micro plasma for treating pollutant gases, since ozone is one of the strongest oxidizing agents and plays an important role in plasma gaseous reaction [12, 13]. Other active species such as O, N and OH radicals are also playing a key role to control pollutants in air.

Fig. 7, 8 shows the characteristics of ozone and NOx generation by micro plasma in air. Ozone and NOx concentration are measured 3 minutes after the discharge start considering the response time of each gas analyzer. In this paper, NOx is defined as the sum of NO and NO_2. The air flow rate is set to 8.5 L/min and 4 pairs of electrodes were used.

The generation of ozone increases with increasing number of electrode pairs, although it has a generation peak at lower discharge voltages.

With extra voltage, the reduced electric field, which could generate high energy electrons, becomes high enough to dissociate nitrogen, and to form NO. Generated NO is immediately reacted with generated ozone to form NO_2. The dissociation energy of nitrogen is known to be around 9 eV and the dissociation energy for oxygen is around 5-6 eV. NOx concentration increases linearly with increasing discharge voltages and could be reacting with ozone. There are many reactions occurring in the micro plasma, and the main chemical equations especially related to the generation of NOx are known as the followings [14].

$$N^* + O_3 \rightarrow NO + O_2 \quad (1)$$
$$NO + O_3 \rightarrow NO_2 + O_2 \quad (2)$$

It is able to obtain more ozone with less NOx generation with increasing number of electrodes than by using a single pair, for dry air as the fed gas.

V. Removal of NOx in Dry Air and Nitrogen

By using a micro plasma electrode, we have investigated removal of low concentration NOx. Initial concentration of about 5ppm which is 100 times higher than the EPA guide line [15] is used to estimate the effect of micro plasma.

In this experiment, number of electrode pair is one and gas cylinders are used to obtain the gas composition shown below. Gas flow rate is set at 6 L/min for these series of experiment. Gas Composition: 1) NO 5ppm, nitrogen 80%, oxygen 20%, 2) NO 5ppm, nitrogen 100%.

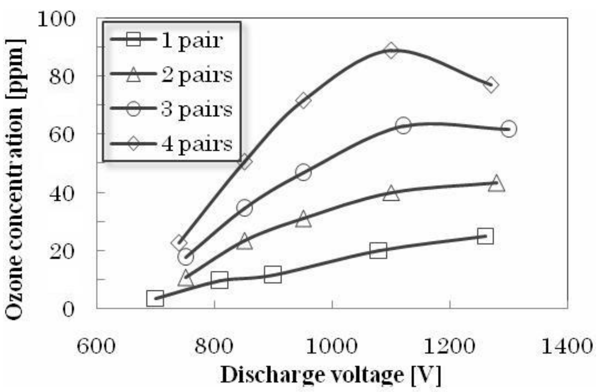


Fig. 7. Generated ozone concentration for various numbers of electrode pair with air flow rate of 8.5 L/min.

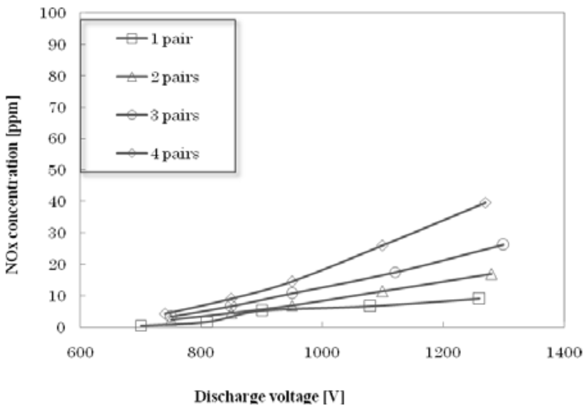


Fig. 8. Generated NOx concentration for various numbers of electrode pairs with air flow rate of 8.5 L/min.

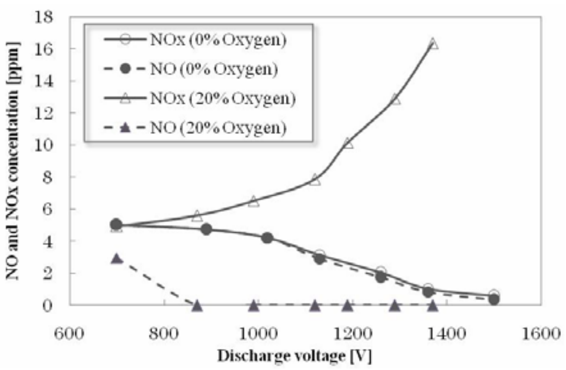


Fig. 9. Low concentration NOx removal by micro plasma electrode with gas flow rate of 6 L/min.

NO is immediately oxidized by applying voltage to NO_2 with oxygen concentration of 20%. In this case, NO_2 concentration is increased to generate NOx. Ozone concentration is also increased already shown in Fig. 7 and Fig. 8. From these characteristics, micro plasma could have enough power to oxidize or decompose low concentration of harmful gases which exist in indoor air. It is considered that the discharge voltage should be kept less than 1 kV to prevent surplus NOx generation in the gaseous reaction.

VI. Effect of Micro Plasma for Cigarette Smoke

Cigarette smoke includes over 200 dangerous and effluvium substances [16]. Real cigarette smoke is used to investigate the effect of micro plasma for decomposition of hazardous materials. There are some reports on treating cigarette smoke with non-thermal plasma [17], although there are few reports on researching the smell.

We have confirmed CH_4, CO_2, CO, H_2O and NH_3 as main peaks by using an FT-IR.

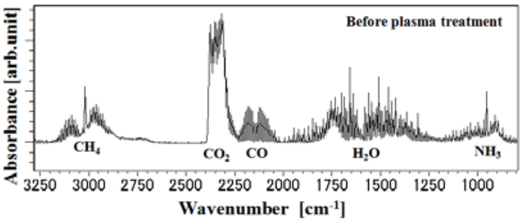
The gas composition of the cigarette smoke and the by-products before and after the treatment is analyzed with an FT-IR and 10 m gas cell, shown in Fig. 10. Decrease of ammonia (920-970 cm-1) is confirmed, which is a major substance of the smell of cigarettes. Nitrogen dioxides (1600 cm-1) and nitrous oxides (2210 cm-1) are found to be a byproduct. The amount of acetaldehyde (2600-2900 cm-1) is small to be detected in this experiment. There are white substances adherent to the plasma electrode after the treatment, which could be ammonium nitrates [18].

It is difficult to specify the concentration change for each major substances of cigarette smoke with the FT-IR, since the amount of water and carbon dioxides are greater compared to other substances in cigarette smoke.

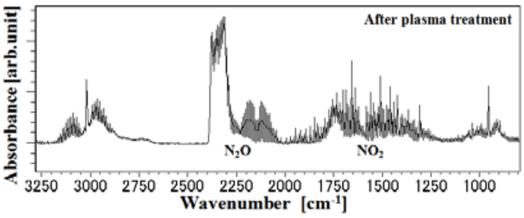
A sensual analysis by a Fragrance Flavor Analyzer is carried out to investigate the feature of the smell of cigarette smoke, before and after micro plasma treatment.

The experimental result is shown in Fig. 11 by comparing the discharge voltage. The experiment is carried out at a discharge voltage of 900 V and a discharge voltage of 1200 V with 4 pairs of electrodes. This data shows the smell similarity between the sample gas and the nine standards gases, which is detected by 10 different types of sensitive oxide semiconductor sensors. Each sensor has a different sensitivity against various substances, and the Fragrance Flavor Analyzer first combines the data from the 10 sensors as a scatter chart for the nine standard gases, which have different types of smell. When the sample gas is analyzed, the measured data is compared with the initial data of the standard gases, and the similarity is calculated.

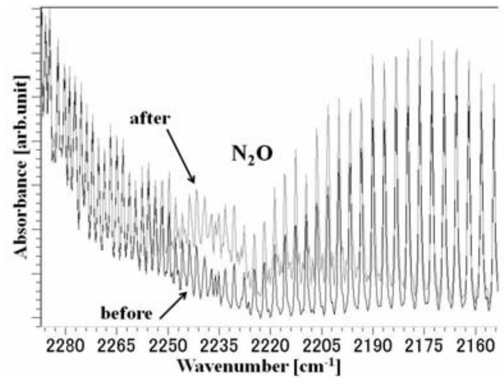
The smell of cigarette smoke has a strong similarity with amine and sulfur. After the plasma treatment, decrease of amine and increase of ester, and aldehyde are confirmed. Oxidation of the amino group has taken place in the treatment. Thus ester and aldehyde could have been formed from oxidation and recombination of amine.



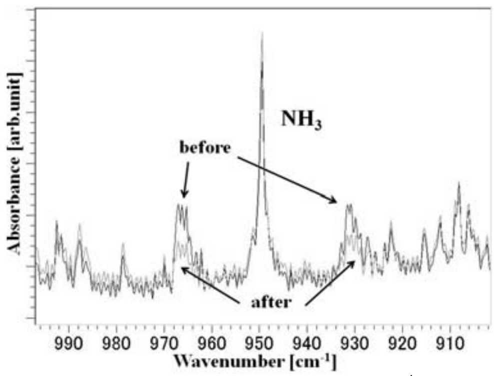
(a) Before treatment by micro plasma



(b) After treatment by micro plasma



(c) Absorbance peaks around 2250[cm^{-1}]



(d) Absorbance peaks around 950[cm^{-1}]

Fig. 10. FT-IR analysis of cigarette smoke treatment at a discharge voltage of 1200V. (15%oxygen + nitrogen balance, gas flow rate 3L/min).

The smell of the cigarette smoke has changed enough for human to disregard.

From these results, smell intensity has been decreased, when the discharge voltage is increased from 900 V (corresponding discharge power of 13W) to 1200 V (corresponding discharge power of 28W).

It is noted that decrease of smell intensity does not necessarily suggest the decrease of concentration of ester, sulfur, aldehyde and amine.

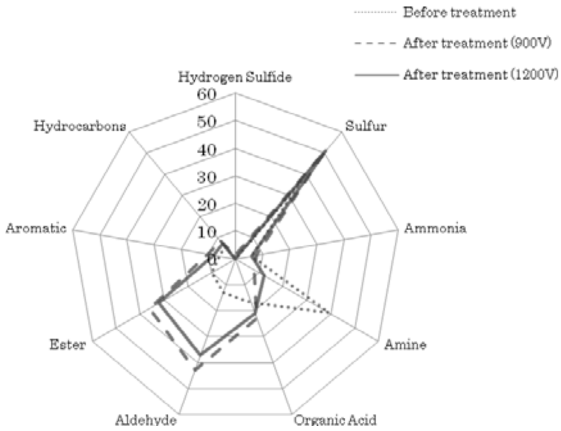


Fig. 11. Similarity percentages against standard gases of cigarette smoke by the Fragrance Flavor Analyzer.

V. Conclusion

In this paper, various experimental results are obtained such as the electric characteristics, ozone generation, and treatment of the smell of cigarette smoke with atmospheric pressure micro plasma.

The followings are the conclusions for the series of experiments.

(1) The practical discharge power is investigated for the micro plasma electrode by using more than one electrode pair. In this case, it is about 6 W at 1080 V for each electrode.
(2) Intense streamers could be occurring throughout the micro plasma electrode, which is confirmed by a thermograph camera. Strong discharge starts at about 1000V and dielectric loss could be the cause for the heat of the electrode.
(3) Ozone generation has certain peaks with various numbers of electrodes; 90 ppm at 1100 V for 4 pairs. Also, with increasing number of electrodes, high ozone concentration can be obtained with lower NOx generation.
(4) Low concentration NOx is removed by micro plasma for indoor air treatment. In this case, discharge voltage should be kept less than 1 kV to prevent generation of surplus NOx.
(5) Treatment of the smell of a cigarette smoke is carried out by micro plasma. Nitrogen dioxides and nitrous oxides were found as byproducts with FT-IR analysis and decrease of ammonia was confirmed. The smell of the cigarette smoke has change to a sweet smell which derived from ester and aldehyde, after the treatment.

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