Study of Air Pollution Control by Using Micro Plasma Filter

Kazuo Shimizu, *Member, IEEE*, Takeki Sugiyama, and Manisha N. L. Samaratunge

***Abstract*—An atmospheric micro plasma is generated in micro gap electrodes with a dielectric barrier. Discharge voltage is only around 1 kV, although the electrode gap is less than 100 μm. Ozone is generated through the micro plasma electrode, which removes indoor air pollutants such as formaldehyde and nitric oxides. By making the micro gap between dielectric barrier electrodes smaller, a higher ozone concentration could be obtained.**

***Index Terms*—Indoor air pollutants, microdischarge, micro plasma, ozone.**

I. Introduction

NONTHERMAL plasma is widely used for air pollution control methods such as indoor air purification and exhaust gas treatment [1]–[7]. Nonthermal plasma is usually generated by corona discharge [8], surface discharge [9], and silent discharge (or "barrier discharge") [10], [11] at a voltage of 5–10 kV or higher. We have developed electrodes to generate micro plasma with a gap on the order of a micrometer, and its discharge voltage is only around 1 kV. This discharge voltage is relatively lower than that of corona discharge or atmospheric glow discharge. Furthermore, a decrease in discharge voltage will contribute to a reduction of initial cost of power source. This makes the whole system volume compact, containing the electrodes and power source.

To realize the commercialization of plasma for air pollution control, a decrease not only discharge voltage, but also energy consumption or discharge power dissipation of these electrodes, are required. These are particularly necessary for consumer electronics to control house dust or pollutant gases which cause the sick house syndrome. In this paper, various characteristics of micro plasma are described as a device to generate nonthermal plasma for low energy consumption and low discharge voltage.

Furthermore, the removal of toxic gases such as formaldehyde and nitric oxide and ozone generation are investigated by using a micro plasma device (micro plasma filter).

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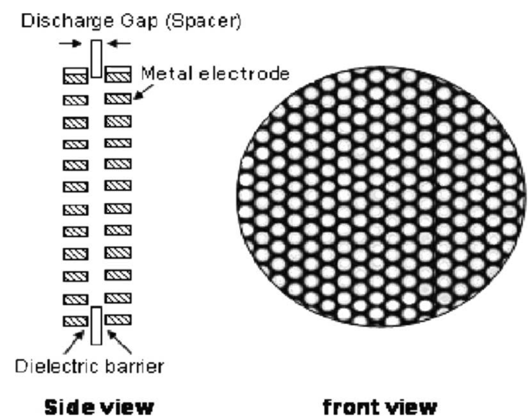

Fig. 1. Structure of dielectric barrier electrodes of micro plasma filter. Processing gas is passed through and purified by a pair of dielectric barrier electrodes which we call the "Micro Plasma Filter." Pressure loss is relatively low even with the narrow discharge gap. The "Micro Plasma Filter" is made of perforated stainless steel (hole diameter is 3 mm, its aperture ratio is 40%, and thickness of electrode is 1 mm) and covered with a dielectric barrier.

TABLE I
EXAMPLE OF MINIMUM SPARKING VOLTAGE AND DISCHARGE GAP AT ATMOSPHERIC PRESSURE

Gas	minimum sparking voltage(V)	minimum gap(um)
Air	330	7.5
Oxygen	450	9.2
Nitrogen	275	9.9

II. About Micro Plasma Filters

A. Dielectric Barrier Electrodes

A pair of perforated metal plates covered with dielectric barrier materials is placed face to face with the discharge gap of 5–100 μm. An alternative voltage at a frequency of 25 kHz is applied to these electrodes to generate nonthermal plasma. Since the discharge gap is smaller than that of the other silent discharge [10], [11], nonthermal plasma is generated. In addition, high energy electrons and active species are generated below 1 kV. Very tense streamers, which generate nonthermal plasma, are observed at the edge of each hole and surface of the dielectric barrier.

Recently, Sakai *et al.* [12] have shown coaxial-hollow microdielectric-barrier-discharges (CM-DBD) to generate micro plasma in small hollow regions. CM-DBD are uniform

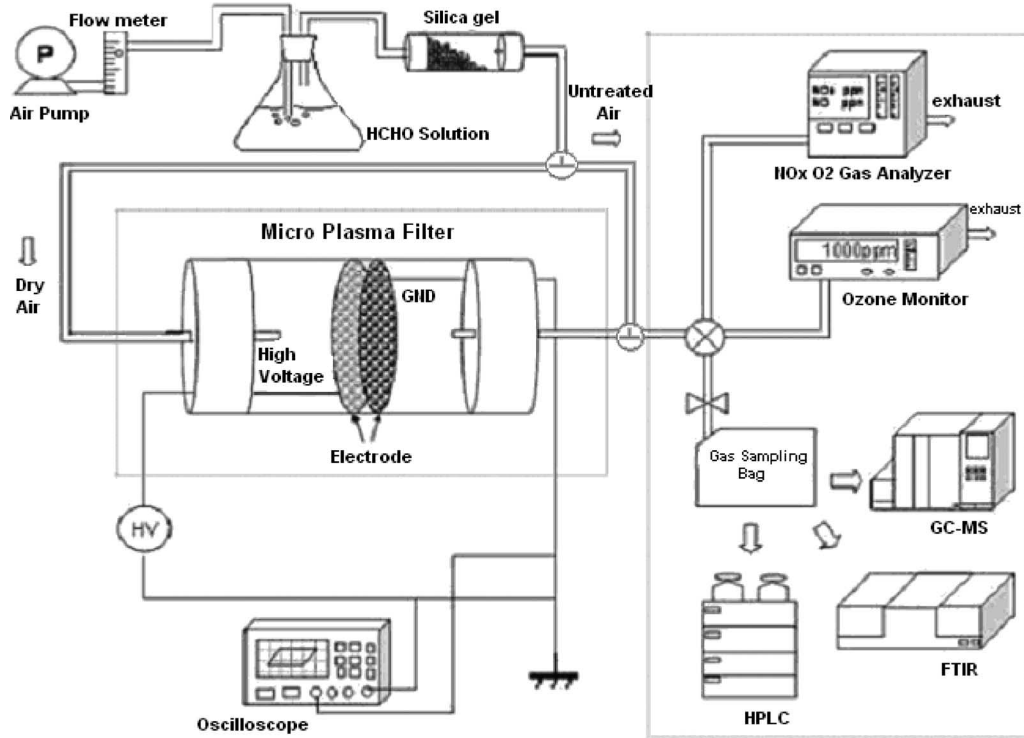

Fig. 2. Experimental setup. Dry air is supplied by using an air pump and silica gel tube. Other gases such as oxygen and nitrogen are supplied from gas cylinders instead of a HCHO solution (not mentioned in the figure). The composition of each gas is quantitatively analyzed by using a GS-MS, HPLC, FTIR, chemiluminescence NO_x analyzer, and ultraviolet detection ozone monitor. Discharge voltage, discharge current, and discharge power are measured by using a high voltage divider, current probe, and digital oscilloscope. Details of electrical measurements are described in the next session/section.

throughout a pressure range less than 2 kV. Geometry of the discharge region and the electrode configuration are different from the micro plasma filter, since CM-DBD do not have a discharge gap between the electrodes.

When processing gas is passed through the electrodes, it is purified since the electrodes work just like a filter. In this paper, this electrode is called micro plasma filter. Pressure loss from these electrodes is less than 5 mm H_2O at a gas flow of 8.5 L/min. An example of the electrode is shown in Fig. 1.

B. Setup for Microdischarge Gap

The discharge gap between electrodes gives a relation between minimum sparking voltage and gas pressure known as Paschen's law. For example, under atmospheric pressure, minimum discharge gap is 7.5 μm for air. Table I [13] shows minimum sparking voltage and discharge gap for various gases.

When discharge gap is set to less than 10 μm and a discharge voltage of 1 kV, a high electrical field $(10^8$ V/m) can be obtained. High energy electrons and active species are generated in the nonthermal plasma activated by a high electrical field. In addition, the population density of low energy electrons (1–2 eV), which cause ozone dissociation, is decreased when the electrical field is high [14], [15].

The ozone is one of the important species and plays a role in nonthermal plasma reaction. The micro plasma filter could be used as an ozonizer when the processing gas is oxygen. It has a potential advantage to produce high ozone density by reducing the population density of such low energy electrons.

III. Experimental Setup

A. Gas Composition Analysis

An odor or toxic gas removal experiment is carried out by using a micro plasma filter. Formaldehyde is regarded as a harmful gas that should be controlled. In Japan, formaldehyde is known as a cause of sick house syndrome and regulated to less than 0.08 ppm in indoor air. Initial concentration of formaldehyde is measured by high-performance liquid chromatography (HPLC) (Agilent Technologies, 1100) and low potential electrolysis sensor (Cosmos, XP-308B). Byproduct generated by decomposition of formaldehyde is analyzed by gas chromatography/mass spectrometry (HP, 5890 Series II) and Fourier transform infrared spectrometer (FTIR) (Shimadzu, IR Prestige-21) with a 10 m gas cell (Infrared analysis, 10-PA). Ozone and NO_x concentration generated by the micro plasma filter is determined by an ozone monitor (Ebara, EG-2001B) based on the principle of ultraviolet absorption and a NO_x analyzer (Shimadzu, NOA-7000) based on the principle of chemiluminescence analysis. The experimental setup is shown in Fig. 2.

B. Power Source for Micro Plasma Filter

Micro plasma does not require a high voltage and can be driven at less than 1 kV. Since the discharge gap is narrow, a high electrical field can be easily obtained with 1 kV. The transformer for neon lighting is utilized to energize micro plasma. Frequency of output voltage is 25 kHz, and this type of transformer is widely used and has a low retail price. Thus,

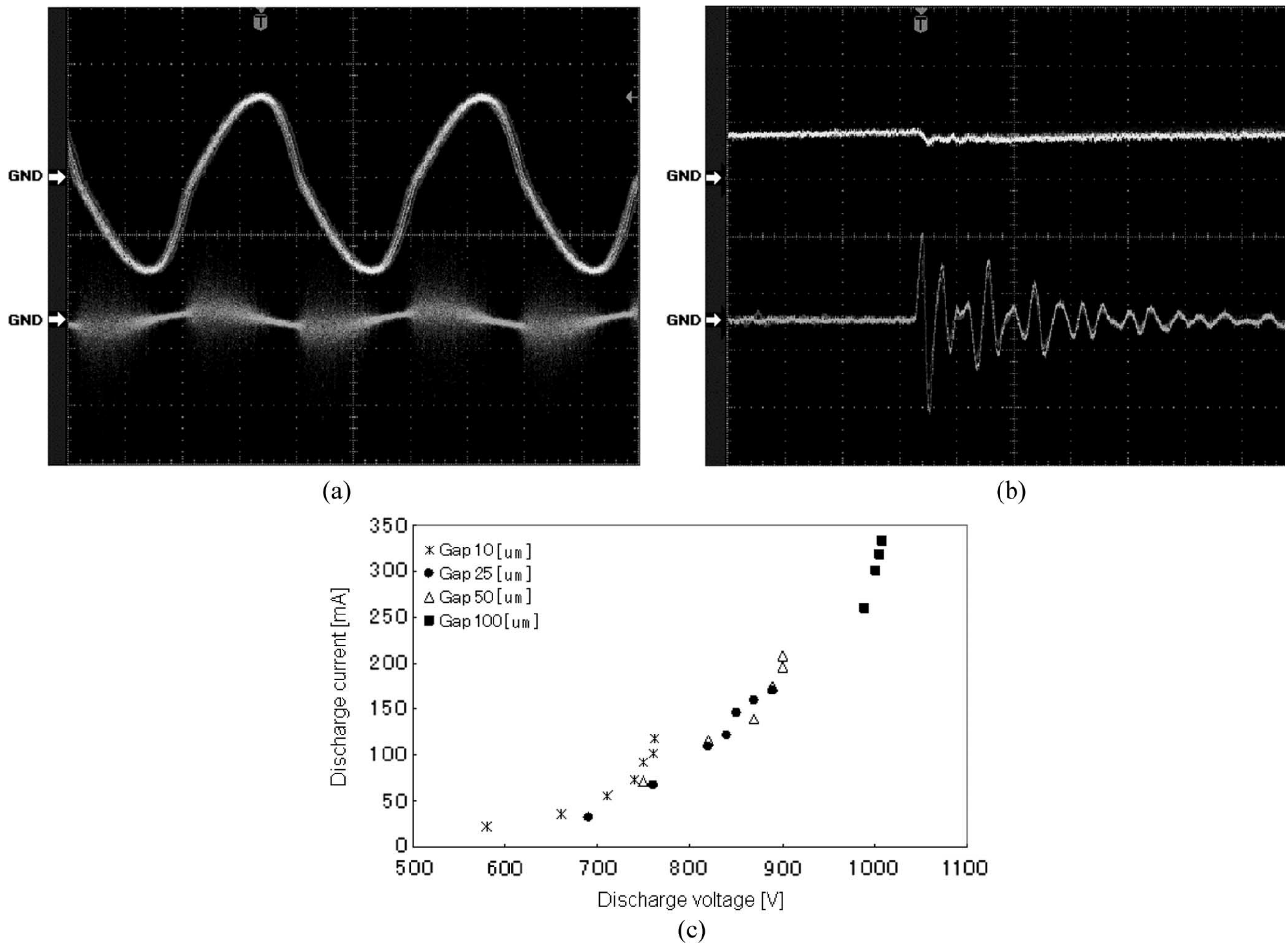

Fig. 3. (a) Discharge voltage and its corresponding discharge current. Discharge voltage: (upper) 1 kV/div. Discharge current: (lower) 200 mA/div. Time scale: 10 μs/div. (b) Discharge voltage and its corresponding discharge current. Discharge voltage: (upper) 1 kV/div. Discharge current: (lower) 500 mA/div. Time scale: 100 ns/div. (c) Voltage and current characteristics of micro plasma filter at various gaps. Note that the streamer discharge is confirmed below 1 kV, while the corresponding discharge current is less than 300 mA. Similar $V-I$ characteristics are obtained in oxygen.

the initial cost for micro plasma is relatively low because of the popular transformer for neon lighting. Fig. 3(a) and (b) show example waveforms of applied voltage and corresponding discharge current waveforms. Oscillated narrow current pulses are observed at both figures.

Fig. 3(c) shows voltage and current characteristics of micro plasma filter in the air. Relatively low discharge voltage and discharge current shown in Fig. 3(c) make discharge power consumption lower than that of the streamer discharge, which has discharge gaps of 100 μm or more. Discharge power is measured by a digital oscilloscope and calculated by a Lissajous figure. Fig. 4 shows an example of the Lissajous figure.

Discharge power is measured at various discharge gaps. When the discharge gap is less than 50 μm, discharge power of 5 W is obtained below discharge voltage of 1 kV. Fig. 5 shows the relation between discharge voltage and discharge power.

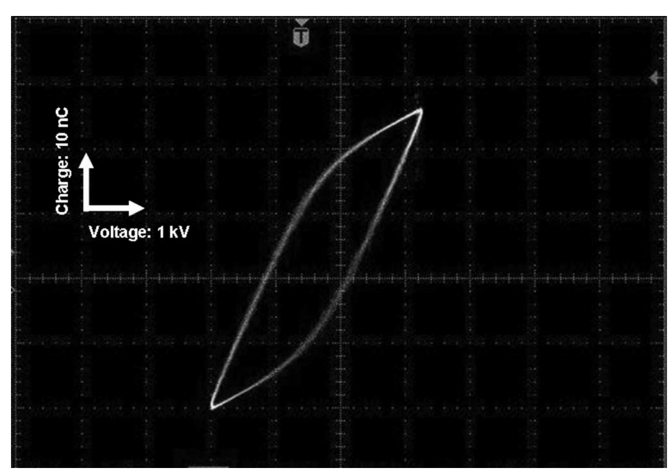

Fig. 4. Area of Lissajous figure corresponds to discharge power of micro plasma filter. x-axis and y-axis of Lissajous figure correspond to discharge voltage and electrical charge, respectively.

IV. Gas Removal Experiment

A. Indoor Air Purification

Indoor air pollutants such as ammonia, formaldehyde, and nitric oxides (NO_x) are strictly regulated by the Ministry of Health, Labor and Welfare in Japan. In this paper, formaldehyde is one of the targets for removal, and its concentration change in micro plasma filter is investigated. Formaldehyde removal with

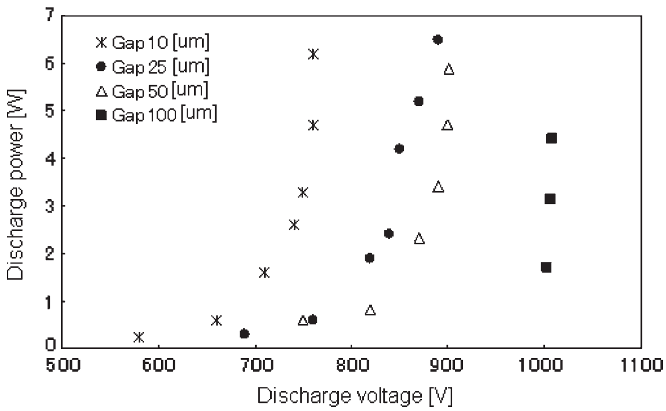

Fig. 5. Discharge voltage versus discharge power. Discharge power increases with discharge voltage, although discharge voltage is regulated by the discharge gap.

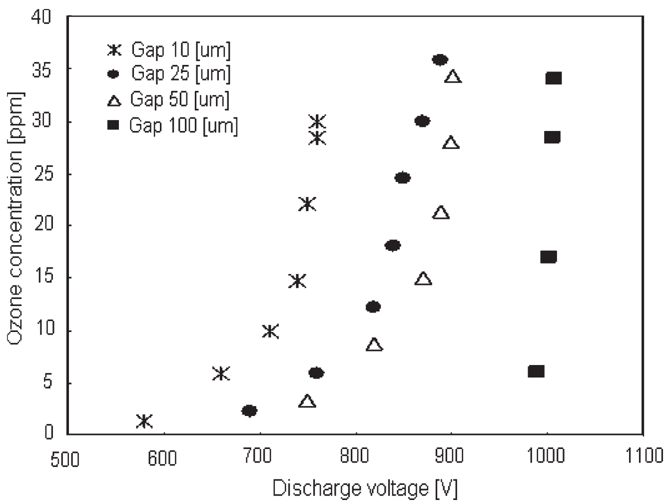

Fig. 6. Ozone generated by micro plasma filter at various gaps. Ozone concentration depends on both discharge voltage and discharge gap between dielectric barrier electrodes. Generation of ozone concentration characteristics are similar to discharge power characteristics already shown in Fig. 5. This means ozone concentration is closely related to discharge power. Dry air is fed to the micro plasma filter at a gas flow rate of 8.5 L/min by using an air pump.

ozone could be achieved by the chemical reaction shown in (1) and (2) and other methods [16]

$$3HCHO + 2O_3 \rightarrow 3H_2O + 3CO_2 \quad (1)$$

$$HCHO + 2O_3 \rightarrow CO_2 + H_2O + 2O_2. \quad (2)$$

The above equations show that ozone plays an important role in decomposing formaldehyde in indoor air. The final products of these reactions are carbon dioxide and water. Ozone concentration is measured in both atmospheric indoor air and pure oxygen by using micro plasma filter. Fig. 6 shows ozone concentration and discharge voltage of micro plasma filter at various discharge gaps.

In this experiment, target gases for removal are formaldehyde and NO_x. The concentrations of both gases are kept low to simulate indoor air. Initial concentration of formaldehyde is set at 4 ppm, although indoor air regulation of formaldehyde is limited to 0.08 ppm by the Ministry of Health, Labor and Welfare. Fig. 7 shows the time course of formaldehyde decomposition by using a micro plasma filter.

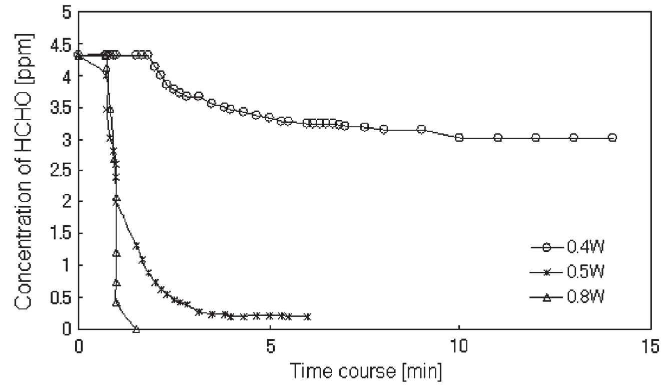

Fig. 7. Time course of formaldehyde concentration by using a micro plasma filter at various discharge powers. Dry air is fed to/into the micro plasma filter at a gas flow rate of 8.5 L/min by using an air pump. Discharge power of 0.4 W is enough for formaldehyde removal, when formaldehyde indoor concentration is several times higher than the regulated level.

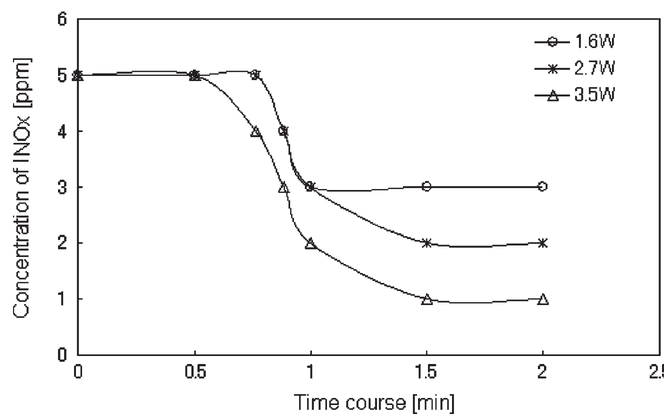

Fig. 8. Time course of NO_x concentration by using a micro plasma filter at various discharge powers. Gas flow rate is set to 8.5 L/min by using gas cylinders. Initial NO_x concentration is set to 5 ppm using NO_x standard gas, and nitrogen is used as background gas. Higher discharge power is required to remove NO_x compared to that consumed removing formaldehyde.

When discharge power is 0.8 W, formaldehyde decreases to zero (below monitor detection level). Indoor air concentration of formaldehyde is two to ten times higher than that of the regulated value in Japan when room walls or furniture are new [17]–[19]. The micro plasma filter removes 1.34 ppm of formaldehyde at a discharge power of 0.4 W. Even when formaldehyde concentration is higher than the regulated indoor level, the filter can remove formaldehyde to below the regulated level at a low discharge power.

It is known that cigarette smoke contains NO_x and other carcinogenic compounds [20], [21]. Recently, smoking has become strictly regulated in public places such as railway stations and schools. With the low level of NO_x now required, the purification method by nonthermal plasma was studied and developed [6], [7].

Fig. 8 shows the time course of NO_x concentration by using a micro plasma filter. NO_x is defined as the summation of nitric oxide and nitric dioxide in this paper. Usually, NO forms about 90% of total NO_x and is oxidized to NO_2 by ozone or other active radicals in nonthermal plasma [22]. Taking into account

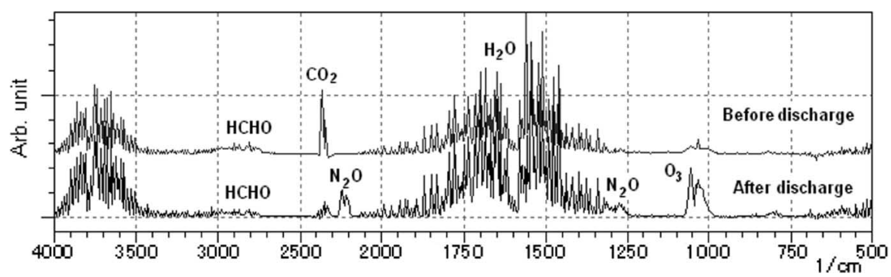

Fig. 9. Byproduct analysis obtained by using FTIR and 10 m gas cell. (Above) Water and carbon dioxide show prominent peaks before discharge. In addition to water and carbon dioxide, (below) ozone and nitrous oxide are found to be final byproducts after plasma treatment. Initial concentration of formaldehyde is 5 ppm, while gas flow rate is 8.5 L/min.

the oxidization process in indoor air, direct NO_x decomposition is hardly achieved. Fig. 8 shows NO_x decreasing at various discharge powers by using a micro plasma filter. Direct NO reduction by $N(^4S)$ could be shown in (3) and (4) [23]–[25]

$$e + N_2 \rightarrow e + N(^4S) + N(^4S,^2D) \tag{3}$$

$$N(^4S) + NO \rightarrow N_2 + O(^3P). \tag{4}$$

B. Byproduct Analysis for Plasma

Some byproducts are found after plasma treatment to remove toxic gases. In the case of treating NO_x with ethylene and oxygen, acetaldehyde and acetic acid emerged as byproducts after plasma treatment [26]. In the conversion of methane to methanol, atmospheric plasma is widely used [27], [28]. In this case, methanol is a byproduct material after the decomposition of methane.

In our experiment, FTIR and 10 m gas cell are used to identify the final byproduct of formaldehyde. Water, carbon dioxide, and oxygen are considered to be final byproducts of decomposing formaldehyde; this is already shown in (1) and (2). However, there is another final byproduct as a result of treating formaldehyde using micro plasma discharge. The FTIR spectrum shown in Fig. 9 suggests that ozone (1054 cm^{-1}) and nitrous oxide (1280 cm^{-1}, 2210 cm^{-1}) are present after micro plasma treatment. The initial concentration of formaldehyde is about 5 ppm, although the corresponding concentration of ozone and nitrous oxide are not determined by FTIR. Since initial concentration of formaldehyde is relatively low to simulate indoor air, other hydrocarbons are not detected by FTIR. Water and carbon dioxide are also found as shown in (1) and (2), as the sensitivity of FTIR measurement to these materials allowed their recognition in the process gas before micro plasma treatment (above spectrum in Fig. 9). These spectra are obstacles to analyzing low concentrations of formaldehyde or other byproducts. Since carbon dioxide is already known to be a byproduct, it was removed from the data to minimize its influence on accurate measurements by FTIR software.

V. Conclusion

Atmospheric micro plasma is generated in micro gap electrodes with a dielectric barrier. Discharge gap is set at less than 100 μm, and its discharge voltage is lower than that of regular corona or atmospheric glow discharge. Successful removal of indoor air pollutants such as formaldehyde and NO_x is carried out by using micro plasma filter. Analysis of byproducts is carried out by using FTIR and 10 m gas cell. The following results are obtained.

1) Both discharge voltage and discharge current are lower compared to other general methods of generating nonthermal plasma such as corona discharge; even an intense streamer is confirmed between the micro gap.
2) Generation of ozone increases with a smaller discharge gap at the same level of discharge voltage.
3) The atmospheric micro plasma decomposes formaldehyde, which is known as an indoor air pollutant, and ozone and nitrous oxide are final byproducts in addition to water and carbon dioxide.

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She has been part of a research team which examines the application of nonthermal micro plasma for indoor air purification and has wide research background regarding reduction of indoor air pollutants such as formaldehyde and NO_x with low energy consumption.

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