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# Indoor air Quality Improvement Using Atmospheric Plasma

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Additional information is available at the end of the chapter

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## 1. Introduction

Air pollution is serious problem for human health and for the environment including causing global warming. An interdisciplinary collection of new studies and findings regarding air pollution was previously published [1]. Governments around the world are managing air quality in their countries for the health of their citizens. The management of air pollution involves understanding sources of air pollution, monitoring contaminants, modeling air quality, performing laboratory experiments, controlling indoor air pollution, and eliminating contaminants through various methods. Research activities are carried out on every aspect of air pollution and its control throughout the world to respond to public concerns.

Another book raised concerns about environmental air pollution including indoor air pollution as well. In recent years, the deterioration of indoor air quality (IAQ) has become a concerning issue [2]. Modern construction methods make the spaces inside buildings more air tight. And, various harmful chemicals are used in building materials. Among the pollutants are volatile organic compounds contained in coated materials and adhesives. While activated charcoal filters used in homes and buildings can remove organic molecules, they have a limited life-time and they must be replaced regularly to maintain good performance.

Another pollutant is particulate matter ranging from sub-micron to few micron meters including dust pollution caused by cigarette smoke, house dusts, and various fungi caused by hot and humid air. Air filters have been used to improve indoor air quality for many years. However, most filters used in homes have poor collection efficiencies for smaller particles (less than 10 micrometer in diameter). This low collection efficiency is a problem, especially for 2<sup>nd</sup> hand smoke, dust and pollen particles [3-5].

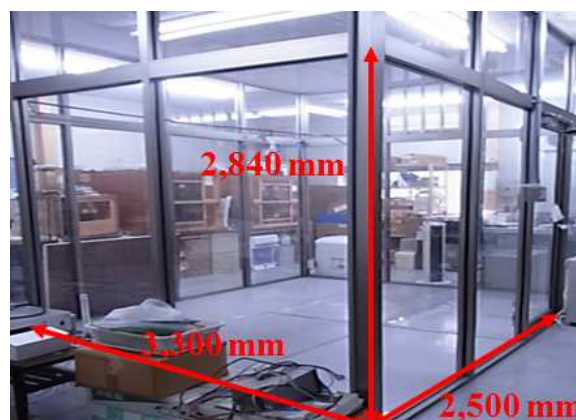
Living microorganisms such as bacteria and viruses also contaminate the indoor air. They may bring on respiratory disorders, asthma, or influenza. And, activated charcoal filters can

incubate bacteria. So the other sterilization method is required for living microorganisms or virus.

Finally, while sterilization devices for indoor air may be commonly used in medical facilities (hospitals, clinics, etc.), these devices are uncommon for use in homes and general office buildings. While these issues could be eliminated by natural air flow, this method is energy inefficient and many houses cannot use this method. Because we spend most of our time indoors, indoor air quality is an important factor for healthy and comfortable lives.

From these points of view, there is a need for reliable devices for the home that can remove organic pollution and that can sterilize indoor air. Devices using atmospheric pressure plasma technologies, especially microplasmas, are very promising.

This chapter deals with improving indoor air quality using atmospheric plasma treatment [6]. Discussed are the results of a series of experiments on particulate matter (PM) precipitation and removal, odor control targeting ammonia and sterilization of *E. coli*. While such experiments are often performed in a small experimental chamber, these experiments were carried out in the relatively large space ( $23.4 \text{ m}^3$ ) shown in Fig. 1. These results reveal the performance of commercially available air treatment devices.



**Figure 1.** The experimental room for measuring indoor air quality improved by atmospheric plasma.

## 2. Experimental set up for PM precipitation

### 2.1. About particulate matter in the indoor air environment

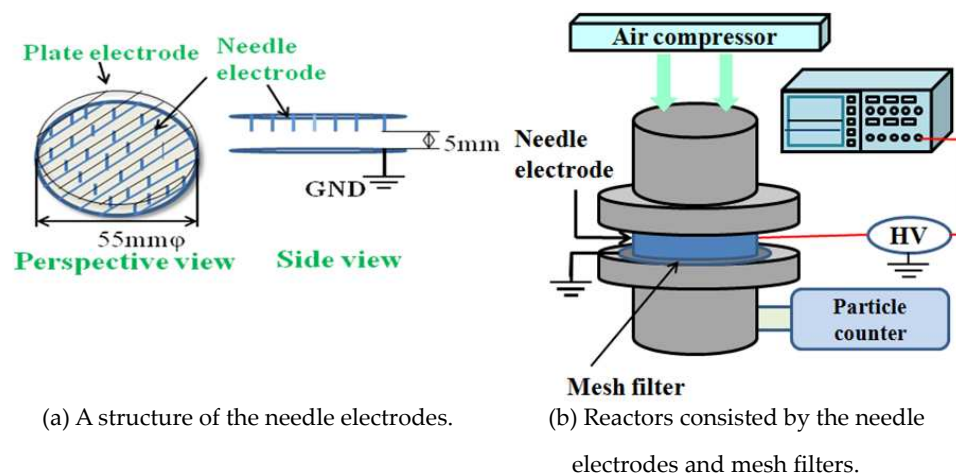
Indoor particulate matter is a mixture of substances including carbon (soot) emitted by combustion sources, tiny liquid or solid particles in aerosols, fungal spores, pollen, and toxins present in bacteria (endotoxins). In many homes, most of the airborne particulate matter comes from the outside. However, some homes do have significant sources of indoor particulate matter that comes from various sources: cigarette smoking is the greatest single source of particulate matter in homes and buildings where people smoke; cooking: especially frying and

sautéing; combustion appliances: for example, furnaces without a proper air filter; non-vented combustion appliances like gas stoves; wood-burning appliances like wood stoves and fireplaces: especially if the smoke leaks or *backdrafts* into the home; and mold growth [7].

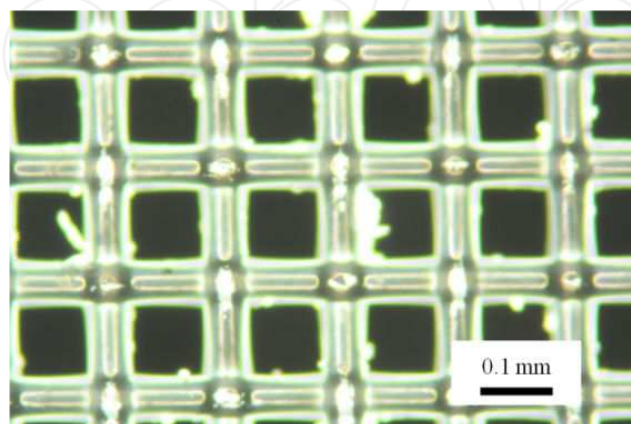
Since indoor air contains various types of particulate matter, improving air quality requires a combination of high performance filters and active treating devices such as electrostatic precipitators and atmospheric plasma devices [8, 9].

## 2.2. Electrodes for particulate matter collection

The reactor shown in Fig. 2 consisted of a corona discharge needle type electrode system, a ground electrode, and a mesh filter electrode for collecting particulate matter. The needle type electrode system shown in Fig. 2 consisted of 5 parallel wires to which 4 needle type electrodes were attached to each wire. The diameter of the electrode assembly was 55 mm. The ground electrode consisted of parallel wires positioned at a discharge gap of about 5mm from needle type electrodes. At the outlet, a mesh filter placed 5mm from grounded electrode captured particles.



**Figure 2.** Schematic diagram of the needle electrodes and the reactor system with the mesh filters.



**Figure 3.** Mesh filter used for collecting dust. (x100)

Mesh filters like the one in Fig. 3 were used in this study. One side of the filters were coated by metal. The characteristics of the mesh filters are given in Table 1.

Aperture size [mm]	0.1
Wire diameter [mm]	0.06
Mesh count [mesh /inch]	150

Table 1. Characteristic of mesh filters.

We used the large, experimental room in Fig. 1 to carry out our indoor air quality experiments using the reactor in Fig. 2. We measured particulate matter collection, and ammonia removal from the indoor air. The fan in the corner of the experimental room shown in Fig. 4 circulated indoor air at a flow rate of 4.6 m<sup>3</sup> / min.

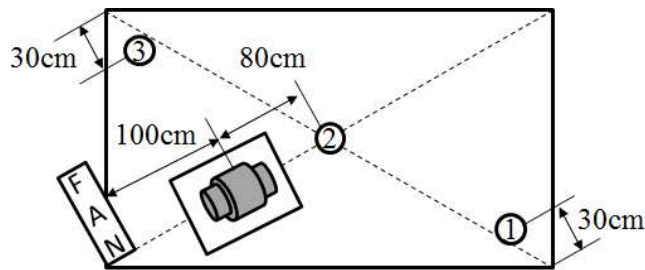


Figure 4. A reactor and a fan arrangement in the experimental room.

The reactor was placed 30 cm above the floor and 100 cm from fan. The gas flow through the reactor was about 112 L / min. The reactor was driven by a negative DC voltage of 5 kV.

To evaluate the collection of particulate matter, we took particle measurements at three locations #1, #2 and #3 in Fig. 4. We measured the number of particles using an optical particle counter (SHIMADZU, MODEL 3886). The diameter of the measured particles was 0.5 μm. During the measurement time of 2 hours, the relative humidity was 67% and the temperature was 23.7 C.

2.3. Microplasma generation for indoor air treatment

The microplasma electrode has been described in previous publications [6, 10]. In this study, the volume of the experimental room shown in Fig. 1 (23.4 m<sup>3</sup>) was much larger than the experimental chambers used in previous studies. So the microplasma electrode in Fig. 5 was enlarged corresponding to the area.

The microplasma electrodes consisted of a high voltage electrode and a grounded electrode separated by a discharge gap in the range 30 to 100 m. Both electrodes were coated with dielectric materials. When an AC voltage in the range from several hundred volts to 1 kV was



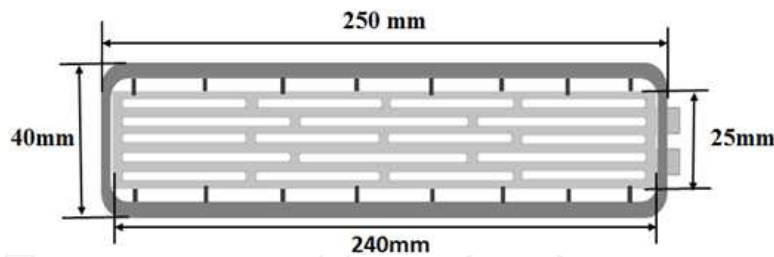


Figure 5. Microplasma electrodes utilized in this study.

applied to the high voltage electrode, numerous streamers were generated between the electrodes as shown in fig. 6 [11].

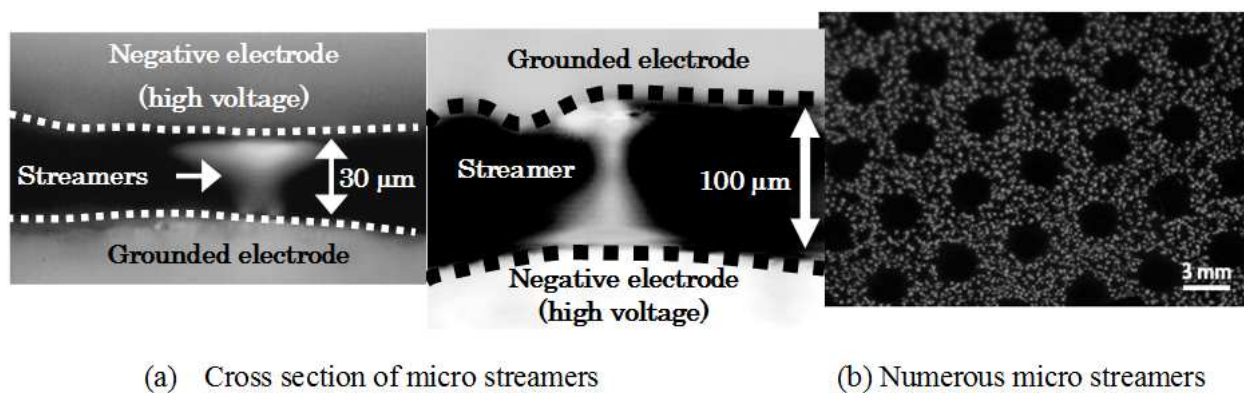


Figure 6. Generation of micro streamers (a); cross section, (b) front view.

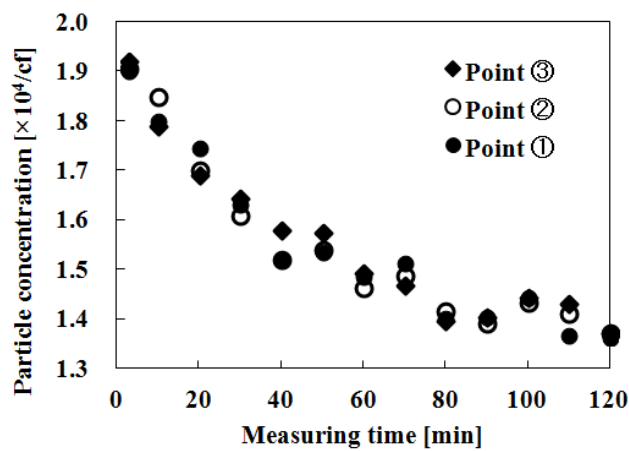
The basic characteristics of a microplasma are similar to that of a typical dielectric barrier discharge (DBD) that typically has a discharge gap of 1mm or more. However, the diameters of streamers in microplasmas were observed to be in the range 10-20  $\mu\text{m}$ , which is narrower than that of DBD streamers [11]. The numerous microplasma streamers generate various active species, such as radicals, ions, and ozone that enhance the chemical reactions in the gas phase describing the next section.

### 3. Results and discussion

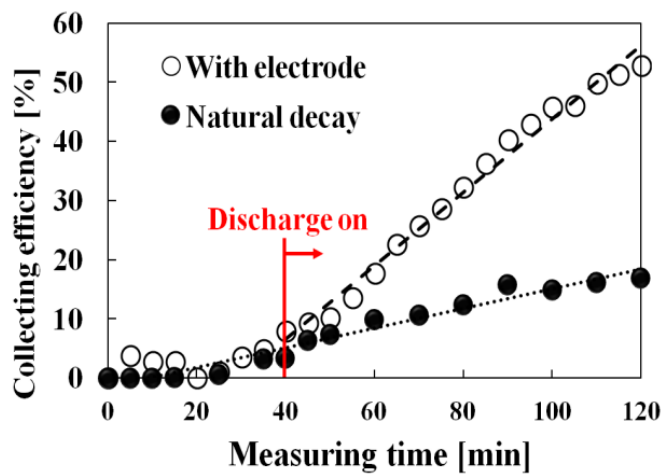
#### 3.1. PM collection by the needle electrodes with the mesh filter

The particle concentrations in Fig. 7 were measured in the experimental room while applying a negative DC voltage of 5 kV.

The measured particle concentrations at the 3 measurement points shown in Fig. 4 were consistent indicating that the air in the experimental room was well mixed by the fan.



**Figure 7.** Particle concentration versus treatment time at 3 measurement points in the experimental room.



**Figure 8.** Decrease of the particles in the experimental room measured at position #2.

Airborne particles will deposit naturally on surfaces resulting in the natural decay of the particle concentration shown in Fig.8. When the power supply of the reactor was turned on for 40 minutes, the deposition of particles on the mesh filter within the reactor increased the decay rate compared to the natural decay (without any discharge). The collection efficiency of particle collection is calculated using equation (1) [12, 13].

$$\eta = \left( 1 - \frac{N_A}{N_B} \right) \times 100(\%) \tag{1}$$

Particulate matter (PM) charged by the needle electrode was then trapped by the mesh filters. The effect of mesh filters and the discharge voltage, the number of filters and the flow rate have been previously published [14].

### 3.2. Odor removal for improving the indoor air quality

This section presents the removal process results from the experimental room ( $23.4 \text{ m}^3$ ) shown in Fig. 1 of chemical substances that deteriorate the indoor air quality. The removal processes target various chemical substances such as volatile organic compounds (VOCs) derived from the building materials, ammonia and others derived from the human being and animals as well. Treatment of VOCs by microplasmas has been published previously [6, 15].

We used the experimental setup shown in Fig. 9 to measure the removal of ammonia from indoor room air. The evaporating dish with 3.5ml of liquid phase ammonia (25%) diffused into the air forming a relatively high initial concentration of ammonia. After confirming the initial concentration of the gas phase, ammonia reached a constant value, the experiment room was ventilated, to evaporate the ammonia again for 10 minutes. Using this procedure, the initial concentration of ammonia was set to 25ppm, which is an acceptable concentration according to the guideline values. Ammonia concentration changes in the experimental room were measured for 2 hours with a negative voltage of 5 kV applied to the needle. The needle electrode was placed in front the fan shown in Fig. 4 at a distance of 100 cm. To enhance the ammonia removal reaction, a fine water mist ( $3 \mu\text{m}$ ) was added by an ultrasonic wave humidifier [16, 17].

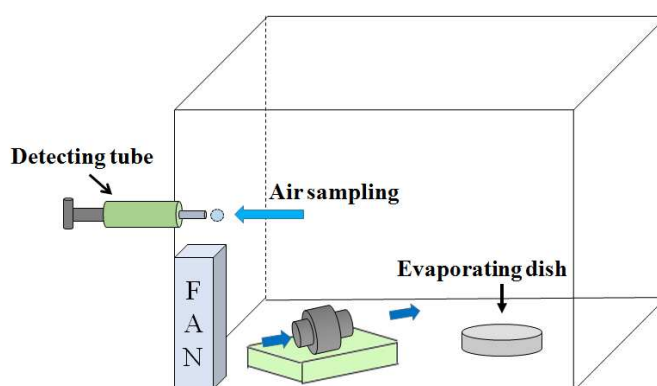


Figure 9. Experimental setup of ammonia removal in the experimental room ( $23.4 \text{ m}^3$ ).

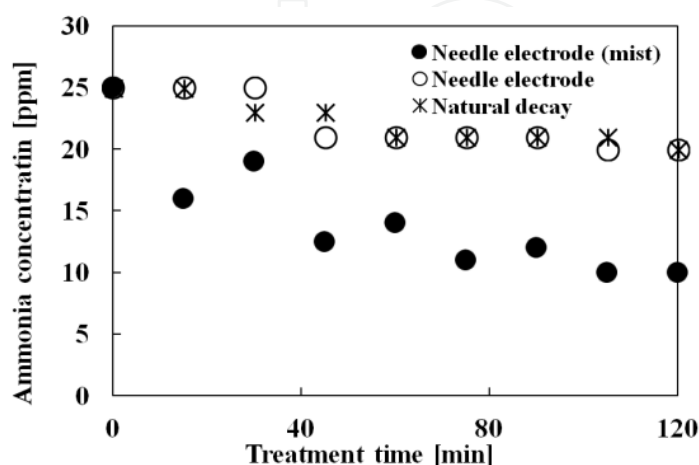
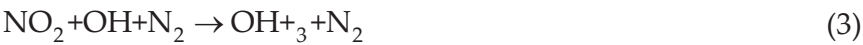


Figure 10. Ammonia concentration versus treatment time by the needle electrode.



During the 2 hours process, the ammonia concentration variation while energizing the needle electrode was the same as the concentration variation due to natural decay. This indicates that the needle electrode removed little ammonia. Note that the ozone concentration in the experimental room after two hours was 0.6 ppm. This relatively low ozone concentration resulted in no measureable reduction in the ammonia concentration. However, when the test was repeated with a fine water mist introduced into the needle electrode, ammonia removal reached 50%.

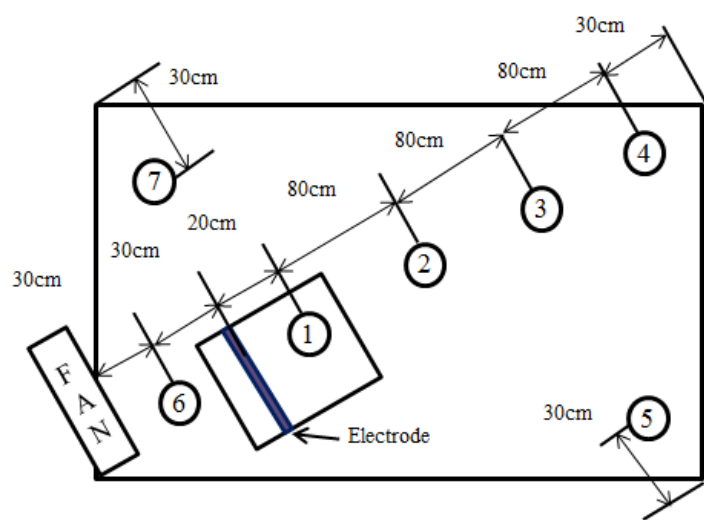
Well known gas phase reactions of ammonia with OH radicals and nitrogen oxides generate ammonium nitrate [18, 19].



These reactions suggest why introducing a fine water mist into the needle electrode greatly increased rate of ammonia removal.

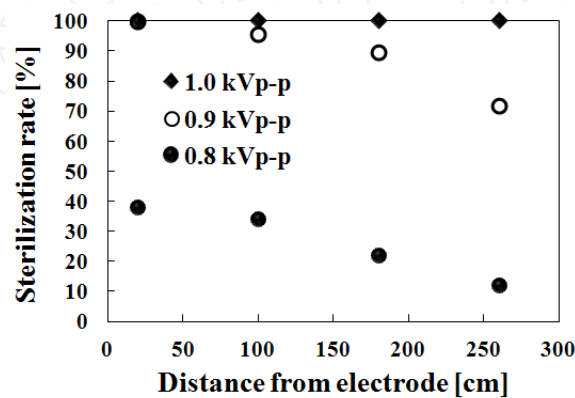
3.3. Sterilization of *E. Coli*

*E. coli* may be sterilized by the various active species formed by a microplasma discharge such as ozone, ions, and radicals [20]. Sterilization of *E. coli* (Migula 1895) was carried out using microplasma electrodes in the experimental room shown in Fig. 1. *E. coli* deposited on stamp media agar “Tricolor (El Mex, Inc.)” were placed at the seven positions shown in Fig. 11 in the experimental room.



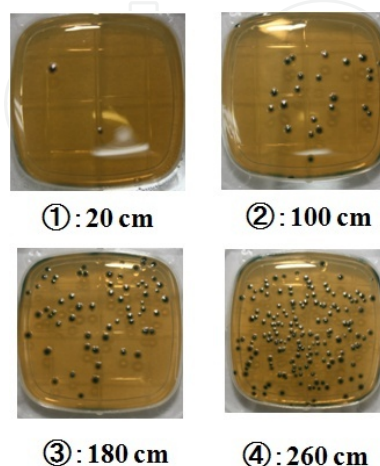
**Figure 11.** Position of the agar media placed in the experimental room.

The target bacteria were placed and treated with the microplasma for 2 hours. An 8<sup>th</sup> control medium with target bacteria was kept outside of the experimental room. Within the room, the microplasma electrodes were driven by a high-frequency alternating voltage of 27 kHz at voltages of 0.8, 0.9 and 1.0 kVp-p by an inverter and a neon transformer. After treatment, the 7 agar media and the 8<sup>th</sup> control medium were cultured for 24 hours in an incubator set to 37 °C. The sterilizing rate was obtained based on the control colonies number compared with the colonies number placed in the each position 1 to 7.



**Figure 12.** Sterilization rate for various discharge voltage at each position in the experimental room.

The sterilization rate increased with increasing voltage and decreased with distance from the electrode with an applied voltage of 0.8 and 0.9 kVp-p. There was no decrease of the sterilization rate with distance from the electrodes when applied voltage was 1.0 kVp-p. Increasing the voltage from 0.8 to 0.9 kVp-p resulted in a considerable increase in the sterilization rate. In the large experimental room, the effect of radicals is likely small since life time of radicals is short compared with the transit time of air moving from the reactor to the agar media test location. Consequently, radicals could not reach the target to sterilize [21, 22]. Figure 13 also shows the plasma treated *E. coli* for various distances from the electrode at applied voltage of 0.9 kVp-p.



**Figure 13.** Plasma treated *E. coli* samples (Applied voltage 0.9 kVp-p).

We also measured the ozone and ions concentration at each position in Fig. 11 in the experimental room. Figure 14 shows the ozone and ions concentration of each position from the electrode, respectively. While the ozone concentration was very high at the outlet of the microplasma electrode, it decreased within a very short distance to a relatively uniform level that varied with voltage. On the contrary, ion concentrations varied strongly with position. This variation of ion concentrations with position was considerably different than the small chamber results. Ion concentrations depended on the distance from the microplasma electrode because ions have various reaction rate, and high reaction rate ions decayed with the distance by reacting with neutral molecules [23, 24].

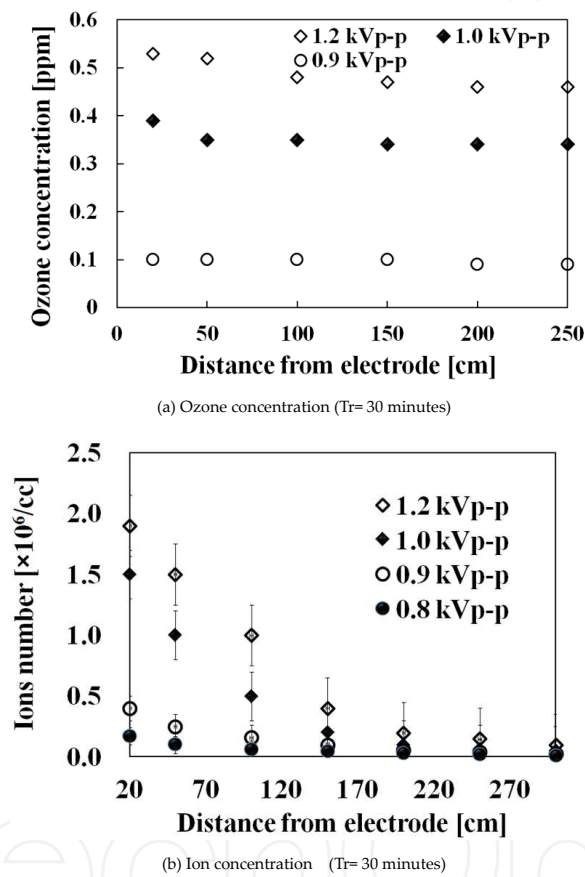
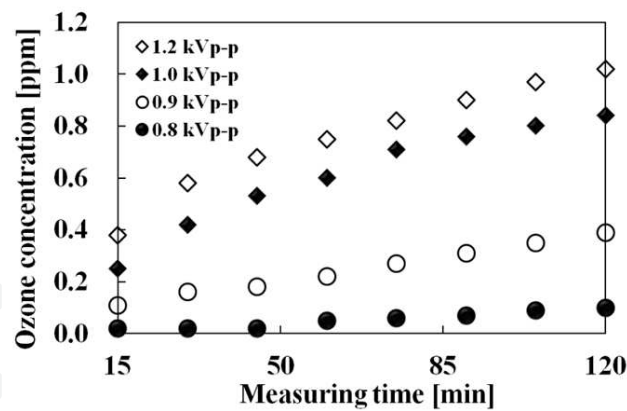


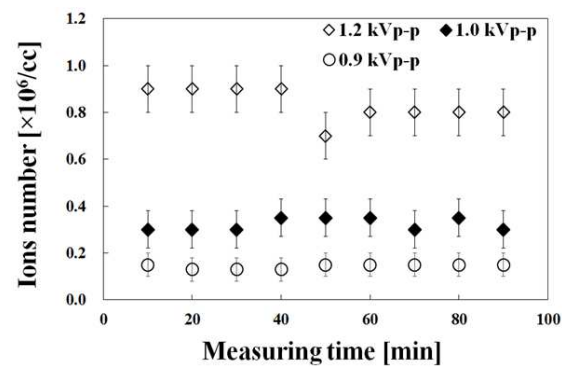
Figure 14. Ozone and ion concentration for various positions in the experimental room.

Even low concentrations of ozone in actual rooms could arise health issues [25, 26], so we investigated both ozone and ion concentration in the large experimental room. The variations of both ozone and ion concentrations are shown in Fig. 15. Ozone concentration increased with the process time, and exceeded the EPA regulatory [27] when the applied voltage was 0.9 kVp-p or higher. The concentrations of ions stabilized as shown in Fig. 15 during the process time (2 hours) at a level that depended on the applied voltage level.

The concentrations of ozone and ions shown in figures 14 and 15 suggest that the sterilization rates shown in Fig. 12 were related to the ozone concentration. However, relatively low



(a) Ozone concentration change (Position; 100cm away from electrodes)



(b) Ions concentration change (Position; 100cm away from electrodes)

**Figure 15.** Variations in ozone and ion concentrations with microplasma treatment time in the experimental room.

concentrations of ozone are generally ineffective in sterilizing bacteria [28]. Similarly, the relatively low concentrations of ions diluted in the experimental room are also generally ineffective in sterilizing bacteria. Note that ions are only weakly reactive compared to highly reactive radicals. Consequently, there should be some reactions with reactive radicals that usually have short life time, such as OH radicals [21].

The ESR analysis of the agar medium in Fig. 16 with spin trap agent (5-dimethyl-1-pyrroline-N-oxide (DMPO)) indicate that the sterilization process may be the OH regeneration process shown in Fig. 17. Low concentration of ozone that reach the agar surface react as in equations (6) – (9) to generate OH radicals that sterilize the bacteria [29, 30].



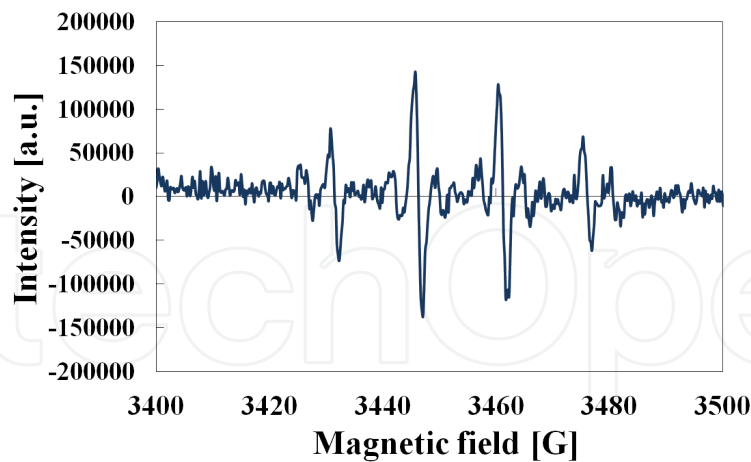


Figure 16. ESR analysis of the medium by plasma exposure.

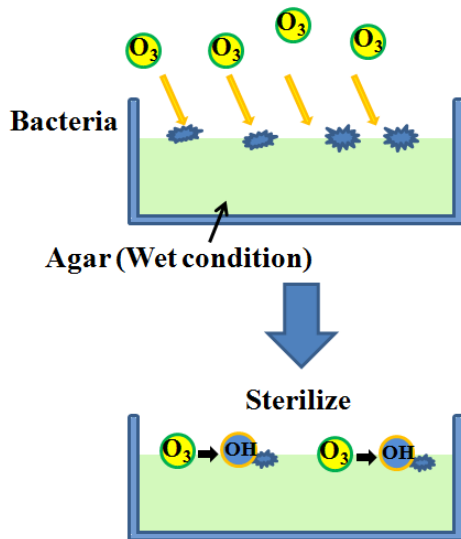
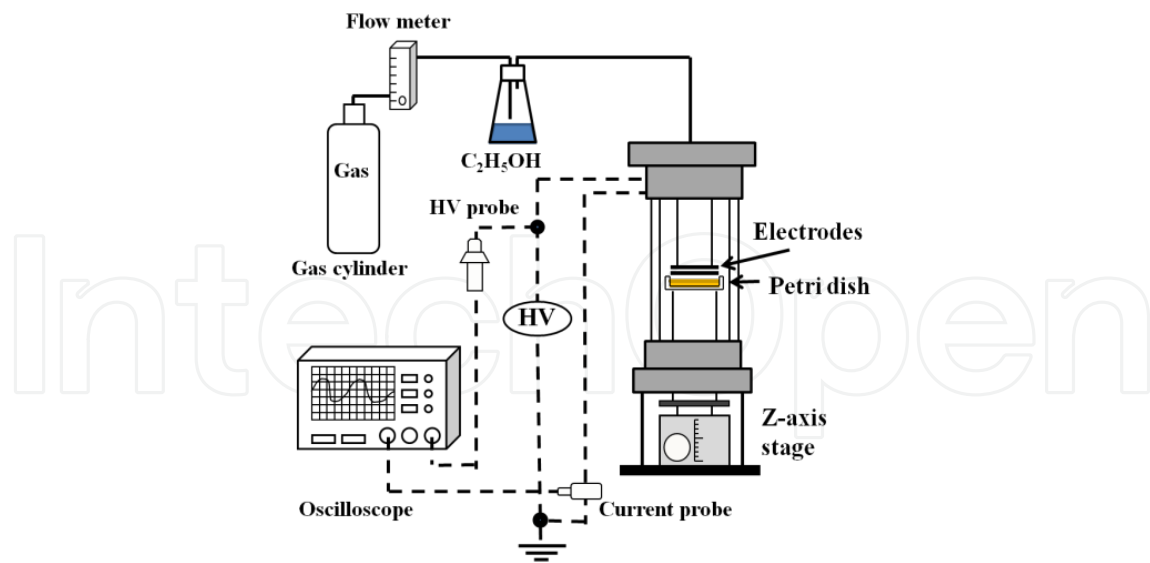


Figure 17. OH generation and sterilization process on the surface of the agar medium.

3.4. Sterilization of *E. coli* with ethanol and microplasma

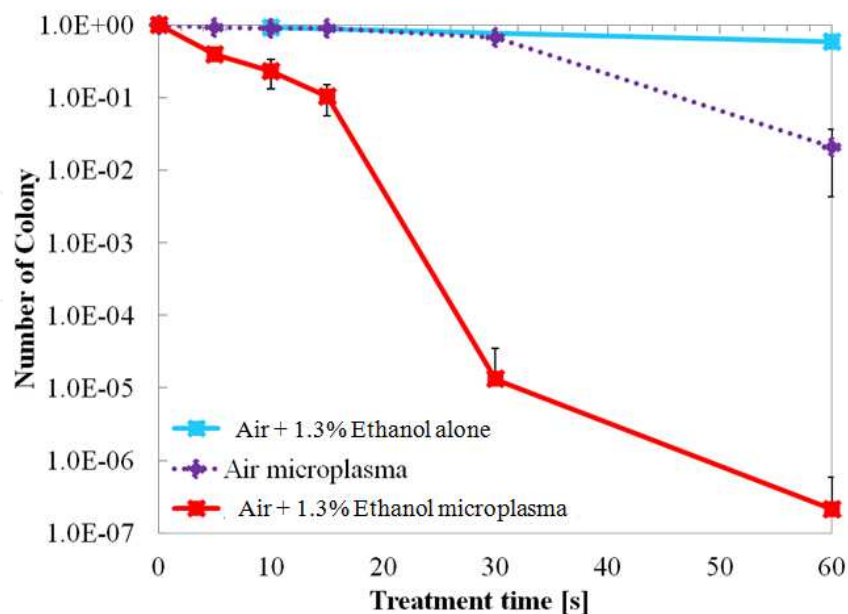
In the large experimental room shown in Fig. 1, *E. coli* was sterilized by  $O_3$  generated by a microplasma rather than by OH radicals whose short lifetime made them ineffective as a sterilization agent. We have enhanced the sterilization effect of bacteria in the small space experimentally as shown in Fig. 18.

A circular microplasma electrode (Diameter 50 mm) was installed in the experimental chamber having an inner diameter of 65 mm. *E. coli* was cultivated on the agar medium in the Petri dish. Air from the cylinder flowed at 10 L/min through a chamber containing liquid ethanol (70 vol %) that evaporated to generate 1.3% gas phase ethanol. The distance between the microplasma electrode and the Petri dish shown in Fig. 18 was 23 mm.



**Figure 18.** Experimental setup for sterilization of *E. coli* by microplasma with addition of ethanol.

With 1.3% gas phase ethanol, the microplasma sterilization process is greatly improved. Without microplasma treatment and only using ethanol, the decrease of bacteria colonies shown in Fig. 19 was minimal. With air microplasma treatment having no ethanol, the sterilization rate reached to 2digits (about 99%) as previously reported [6]. However, the microplasma treatment with 1.3% ethanol as an additive achieved a sterilization rate to 6digits with a 60 seconds treatment time.



**Figure 19.** Sterilization of *E. coli* by microplasma and addition of ethanol.



For sterilizing *E. coli* in a large room such as the experimental room shown in Fig. 1, the life time of highly reactive radicals is too short for effective performance. Thus, experimental work was done to extend the distance between the microplasma electrode and the Petri dish in the small experimental chamber shown in Fig. 18. We found as shown in Fig. 20 that the sterilization performance of the microplasma with ethanol was effective at more than 20 cm away from the Petri dish. Relatively long life species must be generated by the microplasma with ethanol.

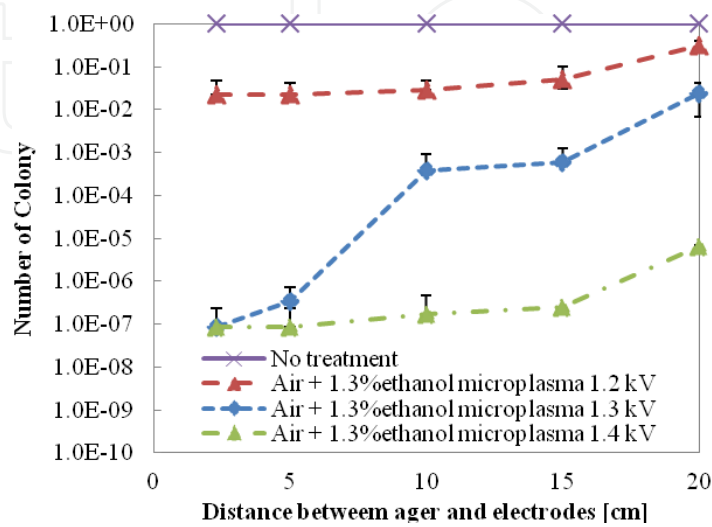


Figure 20. Sterilization rate dependencies with the distance between microplasma electrode and Petri dish.

Byproduct analysis of the process gas was carried out using FT-IR to identify the relatively long life species in the sterilization process. We found the undesirable byproducts shown in Fig. 21 with the ethanol additive process such as CO, and EOG (ethylene oxide gas). EOG is undesirable because it is well known to cause cancer. However, it should be noted that this process would be suitable for applications having a controlled room without any human [31].

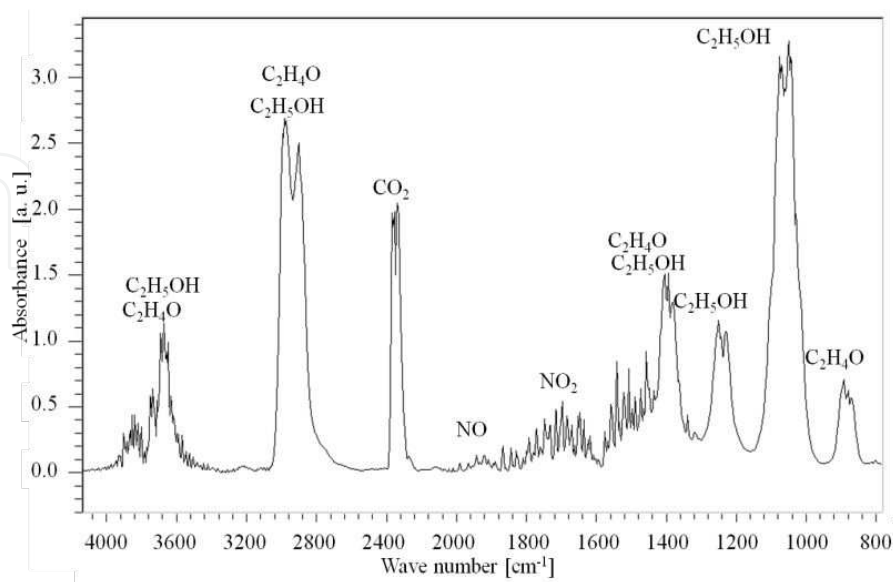
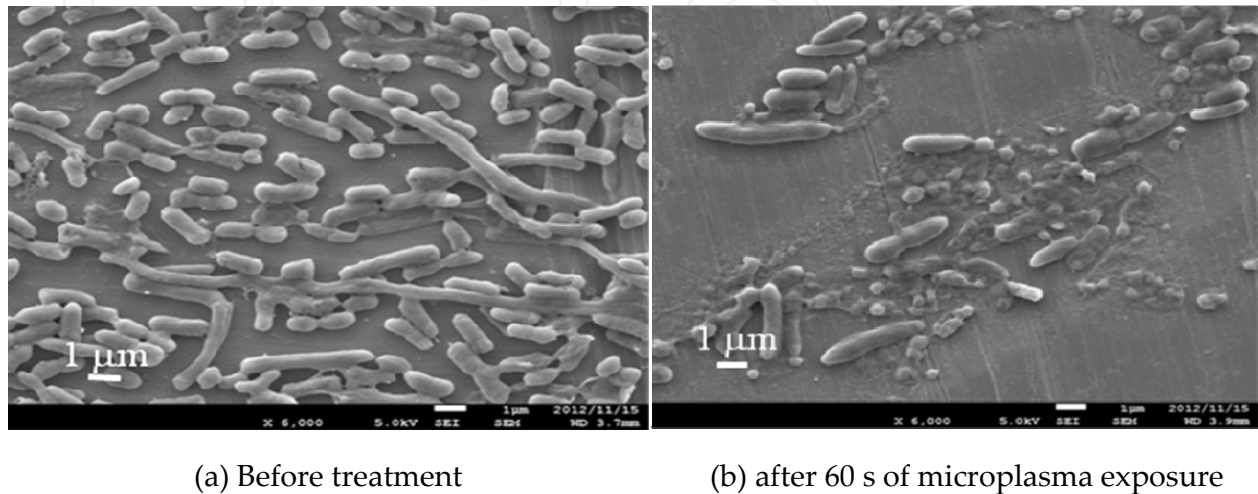


Figure 21. Byproduct analysis by FTIR for the microplasma treatment with addition of ethanol.

The shape of *E. coli* in the SEM images shown in Fig. 22 was completely changed by the microplasma process with ethanol addition. Note that this microplasma process is a remote plasma process. The electric fields and forces acting within the microplasma discharges are not acting on sample. Only various active species or chemical substances generated by the microplasma play a role in sterilizing the *E. coli*.



**Figure 22.** SEM images of microplasma treatment with ethanol addition ( $V_p=1.4$  kV, treatment time; 60 s., ethanol concentration 1.3%,  $\times 6,000$ ).

## 4. Conclusions

Since indoor air quality (IAQ) is so important for our health, indoor air purifiers are commercially available in Japan and in countries around the world. We have investigated the effect of atmospheric plasma including microplasma based on DBD technology both in a small experimental chamber and in a large experimental chamber that is the size of a room. Our results show that a low concentration of ozone played a role in the sterilization process and in decomposing harmful chemical substances such as ammonia. Conducting mesh and electrostatic filters also played a role in collecting particulate material (PM), which are fine particles.

Plasma treatment technologies are relatively new and their performance remains uncertain. Atmospheric plasma can generate ozone various kinds of radicals, ions, and other active species. We propose various plasma treatments for applications to improve the public health. Commercially available plasma devices based on the corona discharge technology and electrostatic precipitation generate low concentrations of ozone. Our results show that ozone at low concentrations is a surprising effective sterilization agent. However, the academic sector has an important responsibility to public health to reveal that plasma processes can also generate harmful byproducts. Our results with investigating plasma technologies to improve the indoor air quality show great promise. It is our pleasure to understand the usefulness of atmospheric microplasmas and their applications

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