Generations and applications of atmospheric pressure glow discharge by integration of microplasmas

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Abstract. Integration of microplasmas enables us to obtain atmospheric pressure discharges with good macroscopic uniformity and various functions. We demonstrate two specific electrodes of microplasmas for generations of dielectric barrier discharges at atmospheric pressure, and integrated microplasmas on large two-dimensional area are sustained with electron density more than $10^{12}$ cm$^{-3}$. Especially “fabric” type electrode has a promising structure to provide various flexible discharge space and plasma processing on winding surface. Integrated microplasmas in array structure will serve as electromagnetic-wave control devices as well as processing tools, and two aspects are discussed in terms of equivalent dielectrics and metals.

1. Introduction
Atmospheric pressure glow discharges (APGDs) have a number of technical advantages over low pressure discharges maintained in a vacuum chamber. Not only technical developments but also scientific researches have been required to obtain large and stable APGDs. A microplasma, which is sustained in a small gas gap less than 1 mm, is suitable for operation of low-discharge voltage at atmospheric pressure due to the Paschen law. If we take enough distance to diffuse radical particles to form uniform density profile, we can obtain two-dimensional homogeneous processing plane by integration of microplasmas.

We have already reported one scheme for such APGDs using integration of microplasmas working at atmospheric pressure [1, 2]. Here we report another type of electrode composed of insulated line conductors, which is referred to as a “fabric-type electrode”; insulated line conductors are “woven” in a similar way to fabrics for cloths and carpets. Such an electrode is completely flexible, and it can form one- to three-dimensional APGD space by imitating various patterns of threads in the fabrics. Using either type of electrodes, we could obtain homogeneous APGDs in helium, nitrogen, and ambient air. In this report, we characterize integrated microplasmas generated in these two types of electrodes.

Such a discharge composed of micropalsmas can be applied to several fields like surface treatment and chemical vapor deposition [3]. We also applied it to generate columnar plasma arrays referred to as “plasma photonic crystal” [4-8], which will lead to novel application to control electromagnetic waves as described in our previous reports. Above electron plasma frequency where dielectric constant is positive but less than unity, we observed an unidirectional band gap in which electromagnetic waves are forbidden to propagate. Under the electron plasma frequency, where dielectric constant is negative,
unique wave modes with very low group velocity are present. Such phenomena are reviewed here briefly.

2. Generations of atmospheric pressure glow discharge

2.1. Two types of electrodes for integrated microplasmas

Coaxial-type discharges were generated inside aligned holes in two adjacent metal electrodes covered by dielectric layers [1], where gap of the electrodes was around 0.3 mm and total thickness of microplasmas was 0.5-1.0 mm. Uniform integration of microplasmas was observed in helium, nitrogen, and air without any macroscopic discharge instabilities at atmospheric pressure. Electron density $n_e$ was measured using millimeter-wave transmittance method as well as metastable helium atomic density by laser absorption spectroscopy [2]. $n_e$ ranged from $10^{12}$ cm$^{-3}$ to $10^{13}$ cm$^{-3}$. This scheme was applied to atmospheric SiO$_2$ film deposition using tetraethoxysilane [3], and uniform thin film was deposited on two-dimensional substrate surface.

We recently developed new type of APGDs by integration of microplasmas in a flexible electrode structure. Using insulated conductor wires and combining them in the lateral and horizontal directions like “weaving” them, we created two-dimensional sheet electrode, which is referred to as “fabric-type electrode” hereafter. The insulation layer works as both insulator between the electrodes and charge accumulation layer. This former function of insulator between the electrodes leads to no requirement of electrode supporter. Since it accumulates charges carried by discharge currents on the insulator layer, discharge scheme is dielectric-barrier type. The insulator is some kind of Teflon [9] with 150 μm in thickness which covers circular conductor wire with 260 μm in diameter. The insulation layer does not suffer from breakdown damage under 1.5 kV alternative-current bias in water. Electrode on one side should be built up by these insulated wires, but the other side can be bare metal wires in some cases. We note that these two functions of the insulated layer and the discharge mechanisms are similar to the electrode configurations reported in Refs. [10] and [11].

![Figure 1](image)

Figure 1. (a) Outlook of fabric-type electrode with schematic of external electric circuit. Inset photograph shows enlarged photograph of partial electrode area. Black lines are insulated wires and shining lines are bare stainless-steel wires. (b) Visible emission from atmospheric-pressure helium discharge at 1.0 kV$_{op}$. (c) Visible emission from atmospheric-pressure ambient-air discharge at 2.0 kV$_{op}$. 
Using this electrode, we successfully obtained uniform APGDs in the macroscopic view in helium, nitrogen, and air at atmospheric pressure, as shown in Fig. 1. In the following experiments, we used the insulated line conductor as high-voltage electrode and bare stainless steel lines with 300 μm in diameter as grounded electrode. There were gaps between wires, and so we can induce gas flows through them to cool the electrodes and to carry radicals created in microplasmas in the gaps. Visible discharge emission image in helium at atmospheric pressure is shown in Fig. 1(b). Microplasmas were generated almost all over the surface of this structure, and the gaps for gas flowing paths were fulfilled with discharge emission. In the case of air at atmospheric pressure, shown in Fig. 1(c), the microplasmas were generated around the contact surfaces between the insulated conductor line and the stainless steel lines. If we increased the applied voltage for plasma generation, emission-vacancy region in the gaps became smaller. We note that the ambient air used here was not dry and included vaporized water naturally present in 40% humidity at 297 K.

These two types of electrodes were compared in terms of ignition voltage. Specific configuration parameters of the coaxial-type electrode were shown in Ref. [1], and we set the thickness of the insulator layer (Al₂O₃) to be 175 μm to avoid breakdown damage. The thickness of the insulator with dielectric constant around 2.1 in the fabric-type electrode was 150 μm, and so equivalent capacitance between electrode conductors in unit area was similar in both cases. Discharge voltage was applied in a square-shaped bipolar pulse with 5 μs width at 5 kHz. In these experiments gas was contained in a vacuum chamber after pumping up to high vacuum level and with no gas flow. Figure 1 displays outlook of fabric-type electrode used in the experiments. The bare stainless wires were in the plane-weave structure with periodical length around 1.2 mm. When we used helium gas, the microplasmas were generated not only around the electrodes but also in the gaps of the line electrodes (around 500 μm in width).

Figure 2 shows ignition voltage in two types of electrodes as a function of helium gas pressure. Here ignition voltage in a partial area indicates the voltage at which a discharge takes place somewhere in the electrode surface, and ignition voltage in the entire area means that the voltage at which a discharge occupies the entire area of the electrode surface. In both electrodes the ignition voltage remains quite low and shows small change throughout this pressure range since the electrode gap is not defined specifically; discharge path for ignition would be shorter when gas pressure increases, and the Paschen minimum voltage in the configuration of parallel-plate electrodes in helium gas lies around these electrode gaps (0.1-1.0 mm) at atmospheric pressure.

![Figure 2. Ignition voltage as a function of helium gas pressure. Experimental data using both fabric-type and coaxial-type electrodes are included. Ignition voltage in “partial” area indicates that discharge takes place somewhere in electrode area. On the other hand, ignition voltage in “entire” area indicates that discharge takes place everywhere on all surface of electrode area.](image-url)
Figure 3 shows ignition voltage in two types of electrodes as a function of nitrogen gas pressure. In both electrodes the ignition voltage increased as the gas pressure was raised. In the nitrogen case, the Paschen minimum will be less than these electrode gaps at atmospheric pressure. The voltage level was comparable or slightly smaller in the case of the fabric type. It is very beneficial to obtain such a low discharge voltage at nitrogen gas which is inexpensive in industry. Figure 4 shows ignition voltage in the fabric type as a function of ambient air gas pressure. The tendency was almost similar to the case in N$_2$, and no gas container is necessary to ignite APGDs using this type of electrode if production of harmful radical species such as ozone is acceptable.

### 2.2. Evaluation of electron density of microplasmas

To characterize these APGDs, we performed electron density $n_e$ measurement using millimeter-wave transmittance method [12]. The coaxial-type electrode had small rectangular apertures, and so it works as Faraday shield of wire-grid polarizer. Millimeter wave is linearly polarized when it is launched from pyramidal horn antenna, and the direction of electric fields was set to penetrate electrode plane. To apply similar technique to the fabric-type electrode, periodical length of bare metal wires on one side was set to be three times larger than the other (1.2 mm), which is a similar configuration to faraday shield, too. As seen in Fig. 5(b), the microplasmas occupied the gaps in the electrode, and so the transmittance of the millimetre-wave signal reflected information of $n_e$ in microplasmas. When we evaluated $n_e$ from reduction of the transmittance signal, we assumed sheet plasma with 500 µm in
thickness, which corresponds to the approximate diameter of the insulated line conductor, and with electron temperature of 0.5 eV. A microplasma in He at atmospheric pressure was ionized via Penning ionization with impurity N₂ and has a density peak in the afterglow phase [2], during which its electron temperature can be estimated to be around 0.5 eV.

Figure 5(a) displays temporal signals of transmittance of millimeter waves with discharge signals in the fabric-type electrode. The gas flow perpendicular to the integration plane was set to be 0.5 litre per minute, and such time evolution was similar to that of the coaxial type [2]. In the case of Fig. 5 (a) where discharge voltage was at 0.6 kV₀p, nₑ had two peaks after discharge current flowed at the rising timing of the voltage pulse. On the other hand, when discharge voltage was at 1.0 kV₀p, nₑ had four peaks since the discharge current flowed at the rising and falling timings of the bipolar voltage pulses.

Figure 6 shows nₑ as a function of discharge voltage. Here nₑ signals at the coaxial-type were shown in comparison [2]. nₑ in the fabric-type electrode was 1-4x10¹² cm⁻³, and was lower than that in the coaxial-type electrode, partly because discharge space in the fabric-type electrode was somewhat open to external space whereas microplasmas in the coaxial-type electrode were confined in the hollow structure. nₑ as a function of discharge voltage in the fabric-type electrode was almost constant for less than 0.8 kV₀p but it increased as the voltage was more than 0.8 kV₀p where the discharge currents started to flow both at the beginning and the end of the voltage pulse.

Around 0.8 kV₀p, two transitions of the discharge state would take place. One is saturation of discharge area. As the voltage increased, the discharge area on the insulated wire, on which charged particles were accumulated, would expand and get saturated when all the surface was covered by the accumulated area. During this time, nₑ might be constant, and increase of discharge voltage and current would contribute to expand the discharge area. After the saturation, excessive voltage would increase nₑ.
The other transition is discharge ignition by accumulated charge. When induced voltage by the accumulated charge on the insulator becomes larger than (“partial”) ignition voltage (around 0.38 kV\textsubscript{0p}), discharges at the falling timing of the voltage pulse takes place. This additional discharge would contribute to \(n_e\) built-up in the gas space before plasma particles extinguish in the afterglow phase. These two transitions might determine \(n_e\) dependence on discharge voltage shown in Fig. 6.

3. Applications of atmospheric pressure glow discharge

The APGDs by integration of microplasmas are inhomogeneous in the microscopic view, but if we set the processing surface apart from the integration plane with the distance more than in-plane periodical length of microplasmas, radical particles created in the microplasmas are distributed uniformly on the substrate surface on the assumption that the radical particles are transported in diffusion flux other than convection flux. Such transport schemes were realized in our previous study of the atmospheric-pressure SiO\textsubscript{2} deposition using the coaxial-type electrode [3]. The fabric-type electrode will expand the applicable field for surface processing to the winding surface which is quite common in automobiles and other large objectives that were difficult to be dealt with at low pressure in a vacuum chamber.

Another example we have examined is functionality for control of electromagnetic waves. Like usual photonic crystals composed of dielectrics, we successfully demonstrated experimentally that two-dimensional array of microplasmas showed photonic bands in the microwave, millimeter wave, and sub-terahertz ranges [4-8]. Due to their spatial controllability, microplasmas with 1-2 mm in diameter were arranged in millimeter periodicity. Since their electron density ranged from \(10^{12}\) cm\(^{-3}\) to \(10^{14}\) cm\(^{-3}\), their electron plasma frequency is 10-100 GHz around which their equivalent dielectric constant gets apart from 1 and significant spatial contrast of dielectric constant between microplasmas and vacuum (gas) space mixture emerges. This structure of microplasmas is similar to photonic crystal which is usually composed of dielectrics. One advantage resulting from use of plasmas is controllability of dielectric constant by changing electron density. Another advantage of microplasma photonic crystals over dielectric photonic crystals is dynamic property which arises from their temporal controllability by external power supply for plasma production. Rapid change of spatial pattern in time will lead to novel device of electromagnetic waves from microwave to terahertz waves.

Figure 7 demonstrates band diagram of columnar plasmas in a square lattice in transverse electric field (TE) mode. This band diagram was derived by the modified plane-wave expansion method, where the dielectric constant was assumed to be in the Drude type [7]. Right axis indicates
corresponding frequency in a case with lattice constant of 2.5 mm. Above the electron plasma frequency which is around 40 GHz in Fig. 7, plasmas behave as dielectric materials, and the contrast of the dielectric constant in a lattice is more obvious as the frequency gets close to the plasma frequency. As a result, the first unidirectional band gap in the Γ-X direction around 70 GHz is wide and the band gaps become narrower as the frequency gets higher. Below the electron plasma frequency, wide flat band region (20-32 GHz) with very low group velocity above the surface (Fano) mode is present.

Band gap is one of the typical and significant phenomena of periodical structure, and plasma photonic crystal can give rise to “dynamic” band gap in equivalent dielectric phase above the electron plasma frequency. The unidirectional band gap as shown in Fig. 7 was verified by our previous experiment [4, 7]. Columnar plasmas in square lattice formed two-dimensional plasma array. The lattice constant ranged 1.5-2.5 mm, and \( n_e \) was around \( 10^{13} \) cm\(^{-3} \) where plasmas were produced in coaxial-microplasma-assisted discharges or in direct-current discharges with ballast resistors. When we transmitted millimeter or sub-terahertz waves to the microplasma array with 6-10 mm in height, we successfully observed a band gap in the Γ-X direction, i.e., stopband for electromagnetic waves where transmittance of the wave decreased down to 18% of its initial value. The frequency position and width were consistent with calculated results on band diagram with parameters suitable to the experimental conditions.

Below the electron plasma frequency, the predicted flat bands are composed of surface-wave modes; their properties are similar to those of surface plasmon polaritons on metals [13]. Although these modes are lossy due to large imaginary wave number, localized fields remind us of near-field optics which have led to several important applications in photon range. Since the flat bands have quite low group velocity, they can intersect dispersion curves with a wide range of different characteristic impedance, and so matching to the transmission lines and the propagating space can be fairly good to transmit wave energy without significant reflection.
4. Summary
To obtain atmospheric pressure glow discharges, we showed integration of microplasmas with good two-dimensional macroscopic uniformity and low ignition voltage. Novel electrode structure composed of insulated line conductor enables us to access to the surface processing on winding surfaces, and various structures of plasma space can be designed for specific applications. Electron density in such fabric-type electrodes is more than $10^{12}\text{ cm}^{-3}$, and a stable glow discharge was observed even in the ambient air at atmospheric pressure. Integration of microplasmas gives rise to novel function of a controller of electromagnetic wave as well as a large-area atmospheric-pressure plasma sheet.

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