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Atmospheric linear-type mixed-gas plasma source with low-particle emission for large area surface treatment

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Abstract

In recent years, atmospheric low-temperature plasma has been extensively utilized for surface treatments. In our laboratory, an atmospheric multi-gas plasma jet source capable of generating low-temperature plasma with various gas species was developed. However, due to the 1 mm diameter of the plasma injection hole in this device, two-dimensional scanning was required to treat large areas. To address this limitation and enable large-area treatment with one-dimensional scanning, this study developed an atmospheric linear-type mixed-gas plasma source. This plasma source features a plasma irradiation slit measuring 1 mm × 349 mm, emitting low-temperature plasma in a gas curtain-like manner. Atmospheric low-temperature plasma was generated using various gases by applying a radio frequency (RF) of 13.56 MHz to the electrodes. The performance of this plasma source was evaluated by measuring water contact angles and the particles emitted. Results indicated an enhanced hydrophilic effect when molecular gases were mixed with argon. Specifically, the contact angle decreased by 42 degrees when nitrogen was mixed with argon, compared to a reduction of 30 degrees with pure argon. Additionally, the uniformity of hydrophilic effect of argon plasma mixed with molecular gases improved with higher RF power. For example, when plasma treatment was performed with 2.0% nitrogen mixed into argon, the dispersion across eight measurement points was 3.62 at 500 W RF power, compared to 35.1 at 200 W. Furthermore, particle emission measurements revealed that the particles from this plasma source complied with the class 100 cleanroom standards commonly used in semiconductor manufacturing. These findings suggest that the linear-type mixed-gas plasma source is suitable for applications in semiconductor manufacturing and other industries that require cleanroom environments.

Keywords: Atmospheric plasma, low-temperature plasma, surface treatment, hydrophilization, particle contamination.

1. Introduction

Recently, atmospheric plasma has been widely utilized in industrial applications, including surface treatment [1], semiconductor manufacturing [2], bacterial disinfection [3], and chemical vapor deposition (CVD) [4]. Atmospheric plasma offers distinct advantages, such as the elimination of vacuum equipment requirements and the ability to generate high-density reactive species. These properties enable high-speed processing. Atmospheric low-temperature plasma, in particular, is widely applied in surface treatment because it does not cause thermal damage to the treated objects. Consequently, various atmospheric low-temperature plasma sources have been developed for surface treatment [5–8]. Plasma treatment methods using these sources are generally categorized into direct plasma treatment [9–11] and remote plasma treatment [1, 12, 13]. In the direct plasma treatment method, discharges are generated between electrodes spaced a few millimeters apart, with treated objects placed directly in the discharge area. This method achieves high treatment effectiveness due to the direct interaction between the object and the plasma discharge. However, it presents two challenges. First, treated objects may sustain damage from the discharge. Second, the method is unsuitable for electrically conductive objects. In contrast, the remote plasma treatment method positions treated objects outside the

discharge area, thereby separating the plasma generation and treatment zones. As a result, treated objects are not exposed to discharge damage, and electrically conductive objects can be processed effectively.

The effects of plasma treatment are primarily attributed to oxygen-derived reactive species, such as ozone and oxygen radicals. For metal treatments, these reactive species clean the material surface by vaporizing and removing organic matter attached to it, thereby enhancing surface hydrophilicity [14, 15]. The types and amounts of reactive species generated depend on the plasma generation gases, making it important to select the appropriate gas for the processed material. Consequently, plasma sources capable of generating plasma with various gas species are highly desirable. In our laboratory, an atmospheric multi-gas plasma jet source was developed [16, 17], capable of producing stable atmospheric low-temperature plasma with various gas species. This source has been applied to surface modification experiments on metals and resins [14, 15]. Notably, plasma treatments using molecular gases, such as nitrogen, oxygen, and carbon dioxide, demonstrated higher treatment efficacy compared to treatments with monoatomic gases, such as argon and helium. Although our plasma jet source is adaptable to a wide range of materials, it is not suitable for large-area treatments due to the 1 mm diameter of the plasma injection hole. As a result, two-dimensional scanning is required for large areas, leading to uneven treatment effects. In industrial applications, large materials, such as 12-inch (304 mm) silicon wafers, require one-dimensional scanning to ensure uniform treatment effects [18].

In this study, a new atmospheric linear-type mixed-gas plasma source was developed for large-area treatment by one-dimensional scanning. To evaluate the treatment effects and cleanliness of this plasma source, experiments were conducted on the hydrophilization of copper plates and the measurement of particles emitted from the plasma source.

2. Experimental

2.1 Atmospheric linear-type mixed-gas plasma source

Figure 1 illustrates the atmospheric linear-type mixed-gas plasma source developed in this study, with its cross-sectional view along the x-axis shown in Fig. 2. This plasma source features an irradiation slit measuring 1 mm × 349 mm, from which plasma is emitted in a gas curtain-like manner. The source is enclosed in an aluminum housing that is electrically grounded, allowing for direct contact.

To generate stable plasma with molecular gases, which require high power input [5, 17], a borosilicate glass plate is positioned as dielectric between the electrodes. Without the dielectric, applying high power often caused plasma generation to become unstable over a large area due to arc discharge concentration [5, 19, 20]. The dielectric prevents the concentration of discharge in a single location even under high power conditions [9, 10]. Consequently, the atmospheric linear-type mixed-gas plasma source can produce stable plasma with molecular gases exhibiting high chemical activity.

Plasma was generated by applying a radio frequency of 13.56 MHz to the electrode within the device. Between the atmospheric linear-type mixed gas plasma source and the radio frequency power supply, a matching box was placed. The impedance of the matching box was adjusted so that reflection waves are 0 W. Since plasma generation got unstable when more than 20 L min^{-1} gas was flowed, flow rate of the plasma generation gas set at 20 L min^{-1}. Remote processing was facilitated as the plasma generation gas passed through the discharge area inside the device and was emitted outside through the slit in the form of a gas curtain. To cool the electrode, ethanol at a concentration of 86% and a temperature of –10 ºC was circulated through the plasma source.

To study plasma generation and determine the maximum rate of molecular gases mixed with argon, experiments were conducted using argon, helium, nitrogen-argon, oxygen-argon, and carbon dioxide-argon mixtures, with 200 W applied to the electrode. In addition, emission spectroscopy measurements of each plasma were performed. In the spectroscopy experiment, a multi channel spectrometer (MAYA2000PRO HC-1, OceanOptics. CO. Ltd.) was placed as shown in Fig. 3.

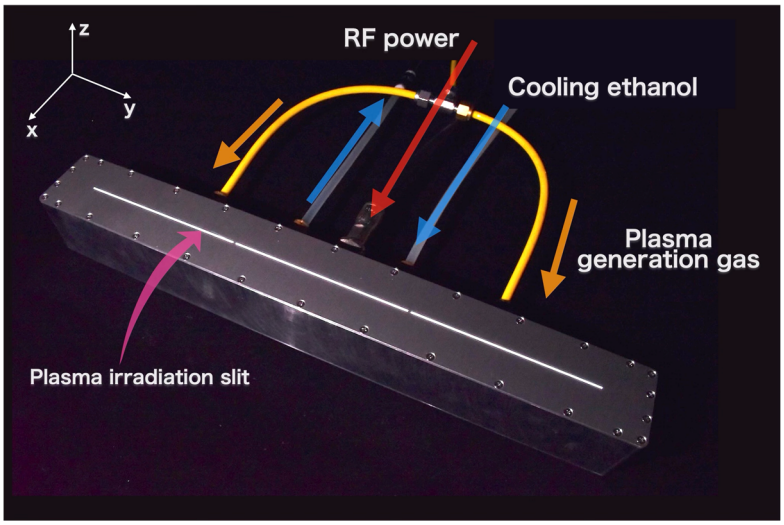

Fig. 1. Atmospheric linear-type mixed-gas plasma source developed in this study.

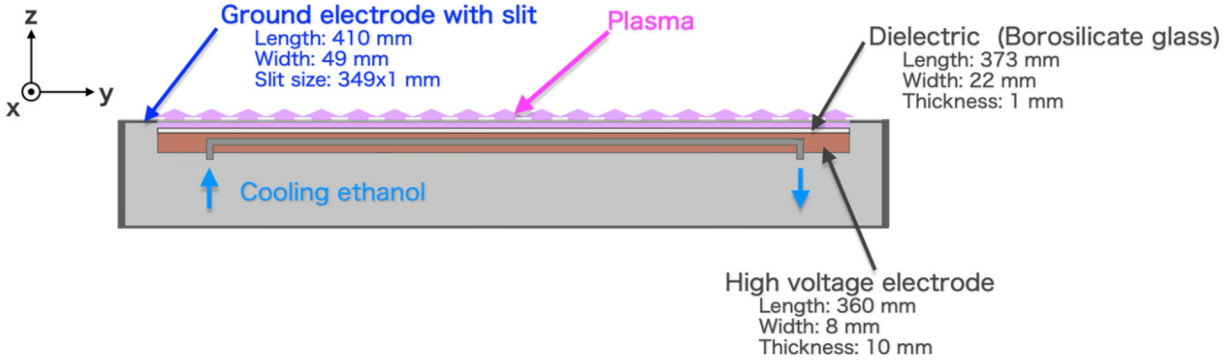

Fig. 2. Cross-sectional view along the x-axis.

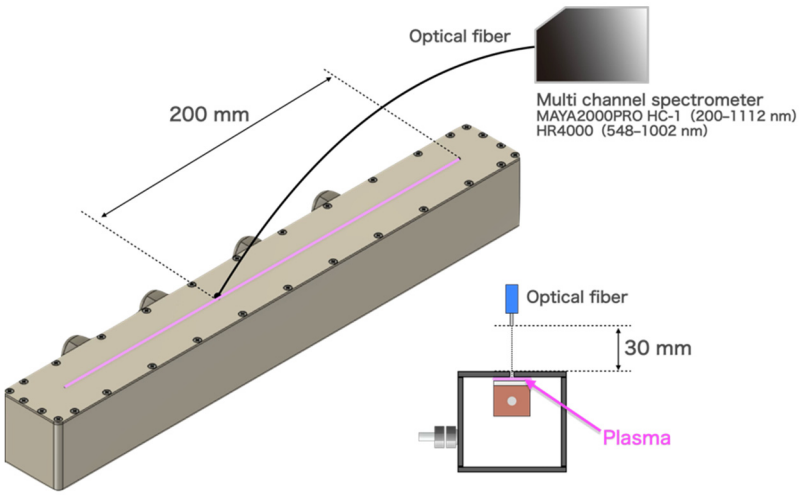

Fig. 3. Emission spectroscopy measurement method of plasma.

2.2 Hydrophilization by plasma irradiation over large area

To evaluate the treatment effects of the atmospheric linear-type mixed-gas plasma source, experiments were conducted on the hydrophilization of copper plates. A schematic diagram of the experimental setup is shown in Fig. 4. Plasma was irradiated to the copper plate at an irradiation distance of 1 mm and a scan speed of 1

mm s^{-1}. Contact angles were measured at eight equally spaced points along the x-axis of the plasma source. Each measurement point had a sample size of five, and the average contact angle at each point was used for evaluation. To examine the effects of gas species and RF power on hydrophilization, the experiments were performed under two different conditions.

First, to investigate the effects of gas species on hydrophilization, treatments were conducted using plasmas generated from argon, helium, nitrogen-argon, oxygen-argon, and carbon dioxide-argon by applying 200 W of RF power to the electrode. The maximum rate of molecular gases that can generate plasma was determined.

Second, to examine the effects of RF power on hydrophilization, treatments were conducted using nitrogen-argon plasma, generated by applying 200 W, 300 W, 400 W, and 500 W of RF power to the electrode.

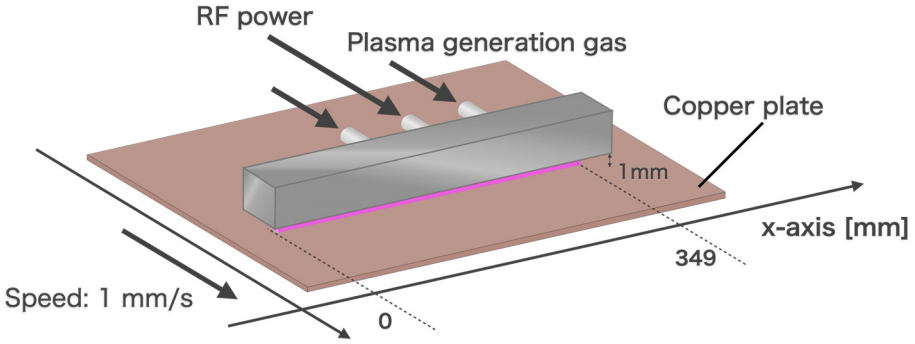

Fig. 4. Schematic diagram of plasma treatment.

2.3 Measurement of emitted particles

In the semiconductor and other manufacturing fields, production takes place in clean rooms where airborne particle levels are controlled to prevent product contamination. Therefore, equipment used in the manufacturing process must also meet clean room standards for particle emissions [21]. To evaluate the cleanliness of the new plasma source, particle emissions from the plasma source were measured. A schematic diagram of the experimental setup is shown in Fig. 5.

In the measurement, the plasma source was placed at deep inside a plastic bag (ASLAB Clean Bags 1-3254-07, AS ONE) of size 460 × 600 mm, and plasma was generated inside the bag. The particles generated were collected from the tip of the vacuum tube of a particle counter (KC-01E, RION Co. Ltd.) placed at the entrance of the bag and the number of particles was measured. Particles were measured in the size ranges of 0.3–0.5 µm, 0.5–1.0 µm, and over 1.0 µm. Five samples were taken, and the average count for each size range was used for evaluation.

For the atmospheric linear-type mixed-gas plasma source, plasmas were generated using argon, helium, nitrogen-argon, oxygen-argon, and carbon dioxide-argon by applying 200 W of RF power to the electrode. The amount of each gas mixture was determined at the maximum rate capable of generating plasma. To compare with a conventional plasma source, the particles emitted from our multi-gas plasma jet source were also measured. The plasma jet was generated using argon at a flow rate of 2 L min^{-1}, applying 9 kV at 16 kHz. The power consumption of the plasma jet was 16 W.

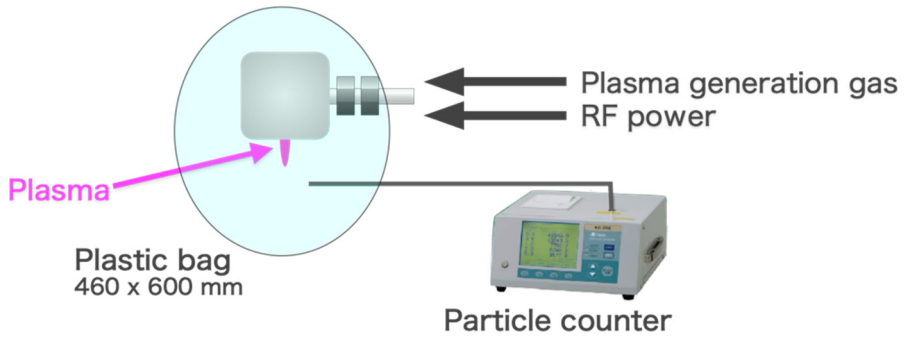

Fig. 5. Schematic diagram of particle measurement.

3. Results and discussion

3.1 Plasma generation by atmospheric linear-type mixed-gas plasma source

As shown in Fig. 6, linear-type plasma was generated using argon, helium, nitrogen-argon, oxygen-argon, and carbon dioxide-argon. The maximum nitrogen concentration for plasma generation at 200 W was 2.0%, while the maximum concentrations for oxygen and carbon dioxide were 0.5%. White light emission was observed from argon and helium plasmas. Notably, when nitrogen was mixed with argon, the plasma color turned reddish-pink. This suggests that the types and amounts of excited species differ when molecular gases are mixed with argon, compared to those generated with argon alone.

The results of the emission spectroscopy measurements of each plasma are shown in Fig. 7. In the argon plasma, emission of hydroxyl radical was observed in addition to argon atomic lines. This is thought to be due to the reaction of plasma and water vapor in the gas cylinder. In the nitrogen-argon plasma, the molecular emission of nitrogen was observed. In the oxygen-argon or carbon dioxide-argon plasma, atomic lines of oxygen were observed.

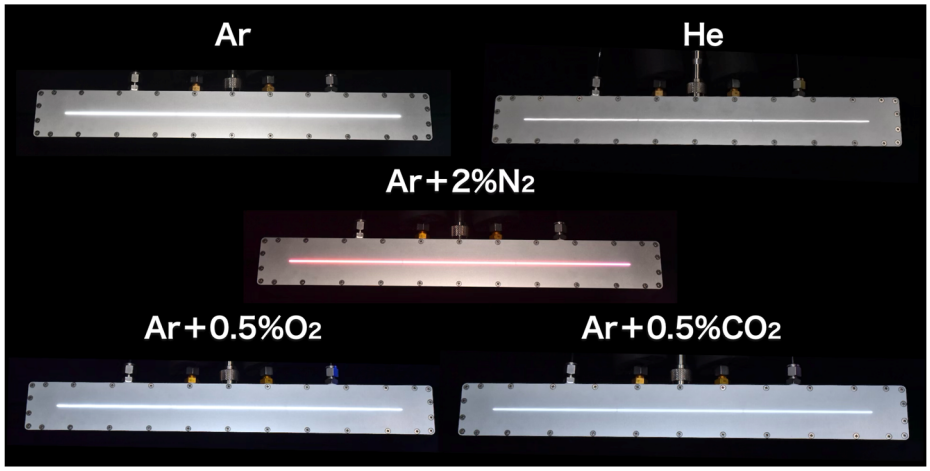

Fig. 6. Plasmas generated by argon, helium, and molecular gases mixed with argon.

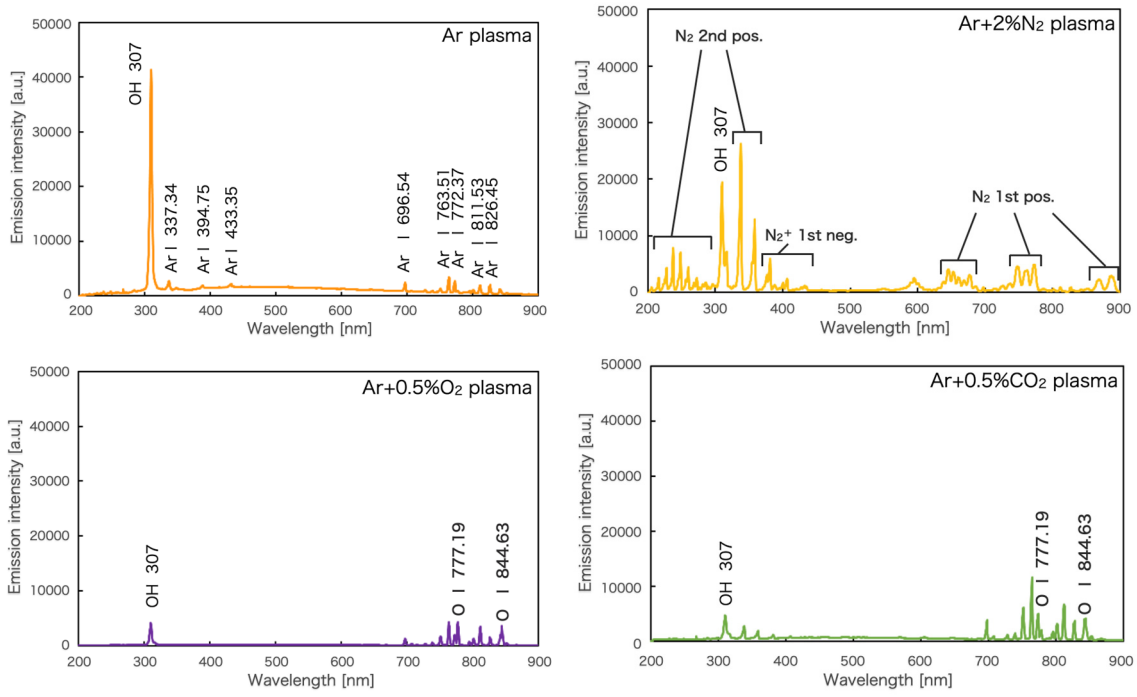

Fig. 7. Emission intensity of each plasma.

3.2 Effect of gas species on hydrophilization

The results of the experiment on the effects of gas species on hydrophilization are shown in Fig. 8, with the averages and dispersions of the contact angles listed in Table 1. The error bars show the dispersion of contact angles at the same position on the slit in the scanning direction of treatment. The initial contact angle was 98 degrees. Hydrophilic effects were observed for all gas species and measurement points. The contact angle decreased to 69 degrees after argon plasma treatment. By mixing molecular gases, the hydrophilic effect was further improved. Notably, when nitrogen was mixed with argon, the contact angle decreased to 56 degrees, indicating a higher hydrophilic effect than with argon plasma alone. Since plasma irradiation distance was close, the hydrophilization effect wasn't affected by humidity of atmosphere. Previous studies [22–24] suggest that oxygen-derived radicals contribute to hydrophilization during plasma treatment, so this result implies that the amounts of oxygen-derived radicals increased when molecular gases were mixed with argon. The mixture of molecular gases generated high chemical activity plasma, leading to the production of oxygen-derived reactive species through reactions with mixed gas or water vapor from the gas cylinder. However, the uniformity of the hydrophilic effect worsened when molecular gases were mixed with argon, compared to argon and helium plasmas, as shown in Table 1. In the case of mixing nitrogen with argon, the dispersion of contact angles at all measurement points was 35.1, 3.8 times higher than that of argon plasma. As mentioned earlier, generating molecular gas plasma requires higher power compared to generating plasma with monoatomic gases [5, 17]. Therefore, we hypothesize that the RF power was insufficient to generate stable plasma with molecular gases over a large area including the edge of the slit.

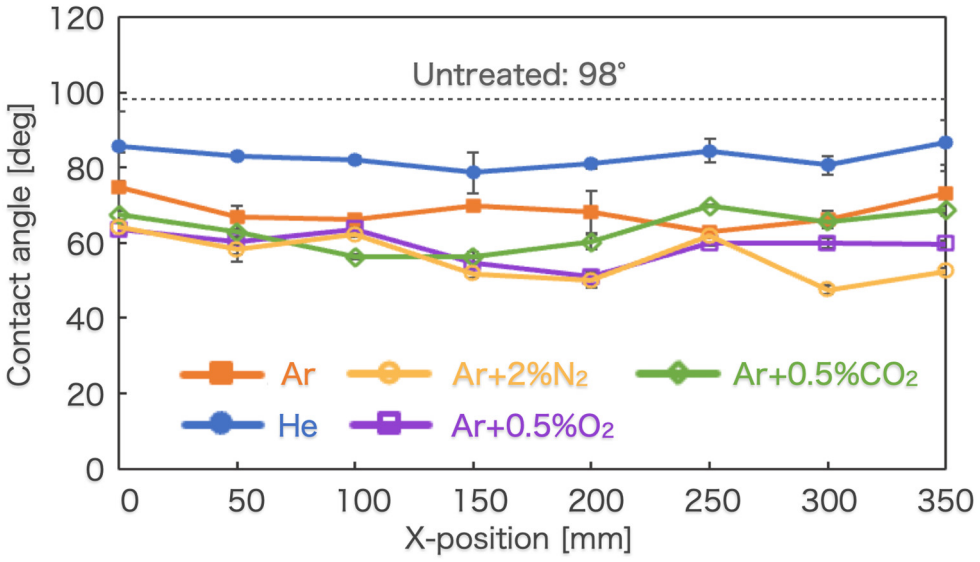

Fig. 8. Hydrophilic effect measured along the x-axis (RF Power: 200 W).

Table 1. Average contact angle and dispersion of measurement points (RF Power: 200 W).

Plasma gas	Average of contact angle [deg]	Dispersion
Ar	68	9.33
He	83	6.48
Ar + 2.0%N_2	56	35.1
Ar + 0.5%O_2	59	18.5
Ar + 0.5%CO_2	63	22.2

3.3 Effect of RF power on hydrophilization

The results of the experiment on the effects of RF power on hydrophilization are shown in Fig. 9, with the averages and dispersions of the contact angles listed in Table 2. The hydrophilic effect improved with higher power. Applying 500 W to the electrodes decreased the contact angle by 10 degrees more than applying 200 W. Notably, the hydrophilic effect improved along the x-axis from 0 mm to 100 mm. At 500 W, the dispersion of the measurement points was 3.62, which was 9.7 times lower than at 200 W. This indicates that the uniformity of the hydrophilic effect improved with increased RF power.

These results suggest that unstable plasma areas existed at lower RF power. In stable plasma areas, nitrogen plasma generated more reactive species than argon plasma [22, 25]. In contrast, in unstable plasma areas, the number of reactive species produced by nitrogen plasma was lower, leading to a reduced hydrophilic effect at lower RF power because of insufficient power supply. Therefore, applying higher RF power supplied sufficient power to generate long plasma and reduced the unstable plasma areas and lowered the dispersion of the measurement points. This indicates that there is an optimal RF power for stable plasma generation when mixing molecular gases, depending on the types and amounts of gas species.

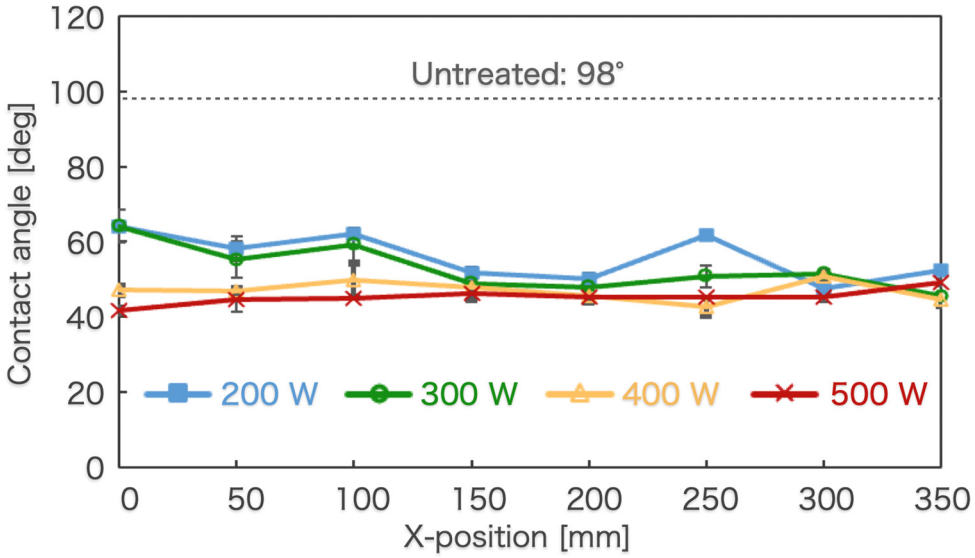

Fig. 9. Hydrophilic effect measured along the x-axis (Plasma gas: Ar+2.0%N_2).

Table 2. Average contact angle and dispersion of measurement points (Plasma gas: Ar+2.0%N_2).

RF Power [W]	Average of contact angle [deg]	Dispersion
200	56	35.1
300	53	34.3
400	47	6.14
500	45	3.62

3.4 Particles emitted from the plasma sources

The results of the experiment on particles emitted from each plasma source are shown in Fig. 8. The dotted line in Fig. 10 indicates the upper limit for the class 100 clean room standard, commonly used in semiconductor manufacturing [26]. For argon plasma generated by the multi-gas plasma jet source, particle counts were 2.0×10^5, 2.8×10^4, and $5.3 \times 1\ 0^3$ counts L^{-1} in the 0.3–0.5 µm, 0.5–1.0 µm and over 1.0 µm ranges respectively. For plasma generated by the linear-type plasma source, argon plasma and helium plasma emitted 7.2, 1.6, and 0 counts L^{-1}, and 6.4, 1.6, and 0 counts L^{-1}, respectively, across each particle size range. Additionally, no particles were detected from the molecular gas-mixed argon plasma in any particle size

range. These results indicate that the particle emissions from all plasmas met the class 100 clean room standard, suggesting that the linear-type mixed-gas plasma source is suitable for semiconductor manufacturing.

These results can be attributed to the frequency of power application. When low-frequency power is applied, electrons and ions are accelerated by the electromagnetic field and collide with the electrodes. This collision causes electrode etching, and the resulting electrode material is thought to have been detected as particles in the experiment using the jet-type plasma source. In contrast, when radio frequency power is applied, ions cannot keep up with the rapid changes in the electromagnetic field and are unable to reach the electrode surface. This reduces the number of ion-electrode collisions, thereby suppressing electrode etching [27]. As a result, the number of particles emitted from the atmospheric linear-type mixed-gas plasma source was lower than that from the multi-gas plasma jet source.

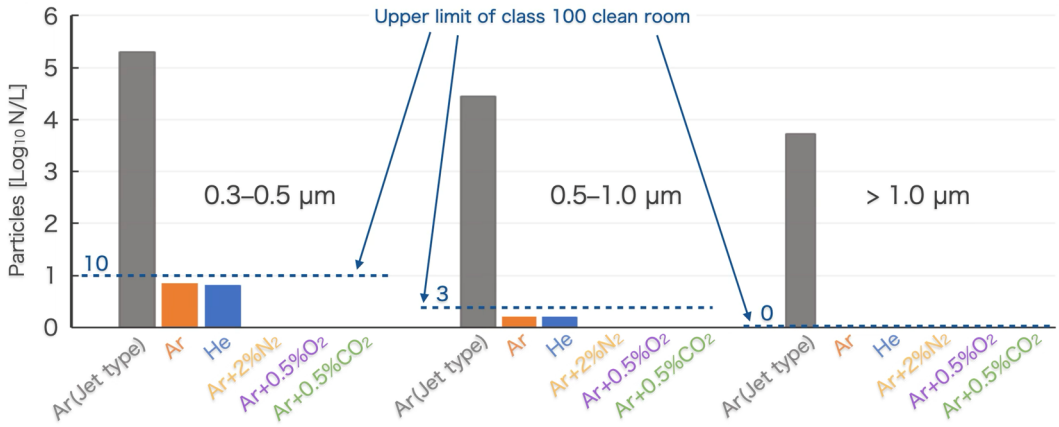

Fig. 10. Particles emitted from the plasma sources.

4. Conclusion

In this study, a new atmospheric linear-type mixed-gas plasma source was developed for large-area treatment by one-dimensional scanning. By applying 200 W of RF power, plasma with argon, helium, nitrogen-argon, oxygen-argon, and carbon dioxide-argon was generated. The maximum nitrogen concentration was 2.0%, while the maximum concentrations of oxygen and carbon dioxide were 0.5% for plasma generation at 200 W. To evaluate the treatment effects and cleanliness of the new plasma source, experiments were conducted on the hydrophilization of copper plates and the measurement of particles emitted from the plasma sources. The hydrophilization results showed that the hydrophilic effect improved with the addition of molecular gases to argon plasma. Furthermore, both the hydrophilic effect and treatment uniformity improved with higher RF power. Particle measurement results indicated that the number of particles emitted from the atmospheric linear-type mixed-gas plasma source met the class 100 clean room standard commonly used in semiconductor manufacturing. Notably, no particles were detected in molecular gas-mixed argon plasma. Our results suggest that mixed-gas plasma treatment provides a higher treatment effect than single-gas plasma treatment. Additionally, the atmospheric linear-type mixed-gas plasma source is expected to be applicable in semiconductor and other manufacturing fields that require clean room conditions.

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