Japanese Journal of Applied Physics
Vol. 44, No. 2, 2005, pp. 1076–1080

Properties and Applications of a Modified Dielectric Barrier Discharge Generated at Atmospheric Pressure

Yong-Hyuk LEE* and Geun-Young YEOM†

Department of Materials Science and Engineering, Sungkyunkwan University, Suwon, Korea

(Received July 7, 2004; accepted November 5, 2004; published February 8, 2005)

An atmospheric pressure plasma was generated using a modified dielectric barrier discharge with the power electrode composed of multi-pins (i.e., a pin-to-plate type) instead of a conventional blank plate (i.e., a DBD-type), and the discharge and the photoresist etching characteristics were compared with those produced by the DBD-type at various He/O_2 mixtures. The pin-to-plate type showed a higher discharge current and a higher power consumption than the DBD-type at a given voltage. Therefore, the pin-to-plate type appeared to be more efficient than the conventional DBD-type. In addition, when the photoresist etch rate was examined, the pin-to-plate showed higher etch rates than the DBD-type at various He/O_2 mixtures. For the He/O_2 mixture, both types showed the maximum photoresist etch rate at a certain He/O_2 mixture. Using a gas mixture of 3 slm of O_2 and 10 slm of He, a maximum photoresist etch rate of 340 nm/min and 260 nm/min could be obtained using the pin-to-plate type and the DBD-type, respectively, at 10 kV AC for an electrode size of 500 mm × 50 mm. No physical damage was observed on the metal lines of the TFT-LCD devices after photoresist etching under the above conditions.
[DOI: 10.1143/JJAP.44.1076]

KEYWORDS: DBD, atmospheric pressure plasma, He/O_2

1. Introduction

Low temperature plasma generated at low pressures is generally used for thin film deposition, surface cleaning, surface modification, dry etching, ion implantation, etc. in semiconductor and flat panel display (FPD) processing. In order to generate the low pressure plasma, costly vacuum generation equipment and measurement tools are required.[1)] In particular, in the next generation FPD processing, the extremely large area of the substrates may make the plasmas generated at low pressure unsuitable to the difficulty and cost in generating a uniform plasma over an extremely large area.

In the case of FPD processing, wet processing is used more than semiconductor processing due to the large dimensions of the device compared with semiconductor devices. However, the use of wet chemical in the next generation flat panel display processing will be restricted due to problems associated with the cost of the chemicals, the environmental impact of the chemical waste, the consistency of the processing, the difficulty of in-line processing, etc in addition to the problems due to the decrease in the device size. Therefore, as a solution to the problems associated with plasma generated in a vacuum and to replace wet chemical processing, large area atmospheric pressure plasma has been actively studied for applications to the FPD processing.[2–5)]

The most widely investigating type of atmospheric pressure plasma for large area processing is a dielectric barrier discharge (DBD). It is composed of two parallel electrodes where one electrode is connected to the alternating current (AC) power supply and the other electrode is connected to the ground. One or two dielectric plates are located either on the powered electrode or on the both of the electrodes between the electrodes. The dielectric plates are needed to limit the current flow and prevent the development of arcing between the electrodes and to generate the plasmas at low voltages by charging the dielectric surface and discharging when the voltage is reversed.[6–9)] This means that a charge develops on the dielectric surface during the half cycle of the sinusoidal AC voltage, which limits the current flow to the electrode, and during the next half cycle of the AC voltage, an electric field develops as a result of the AC voltage and the field is in phase as a result of the charging of the dielectric and the plasma is generated at a lower AC voltage than the voltage required to breakdown the gas at atmospheric pressure.[10,11)] The generated discharge characteristics are dependent on the dielectric materials, the frequency of the AC voltage, the dilution gas, etc.[12,13)]

One of the problems associated with the above conventional DBD-type atmospheric pressure plasmas is the low plasma density. In order to apply the atmospheric pressure plasma to the various low pressure plasma processing and wet chemical processing of the next generation FPD processing, the plasma density, i.e. the processing rates by the atmospheric pressure plasma need to be comparable to those by the low pressure plasma. In this study, as a modification of a typical DBD-type source, a multi pin-type electrode instead of a blank electrode was used as the power electrode to generate the plasma at a lower voltage and to obtain a higher plasma density over a large area, and its plasma and photoresist etch characteristics were compared with the those produced by the typical DBD.

2. Experimental

Figure 1 shows a schematic of the atmospheric pressure plasma system used in this study. As shown in the figure, the top electrode connected to the AC power supply comprised of multi-pins and the bottom ground electrode was a blank plate electrode (pin-to-plate type). In order to operate the discharge as a conventional DBD-type, the top electrode was replaced by a blank plate electrode instead of the multi-pin electrode. The size of the top electrode was 500 mm × 50 mm. On both electrodes, as shown in the figure, 3 mm thick quartz plates were used as the dielectrics in an attempt to limit the current flow. The distance of the air gap between the dielectrics of the electrodes was varied from 2 to 6 mm. A voltage power supply of 3–15 kV (power capacity of

*E-mail address: yhlee73@skku.edu
†E-mail address: gyyeom@skku.edu

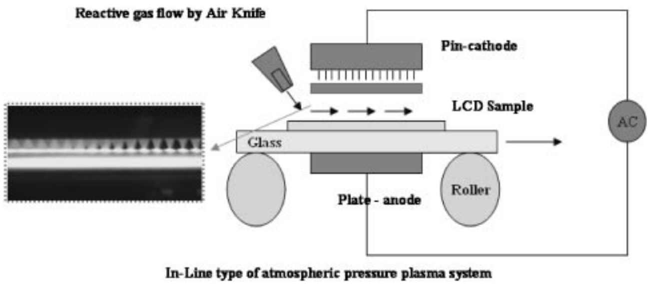


Fig. 1. Schematic diagram of the atmospheric pressure plasma system (pin-to-plate type) used in this study.

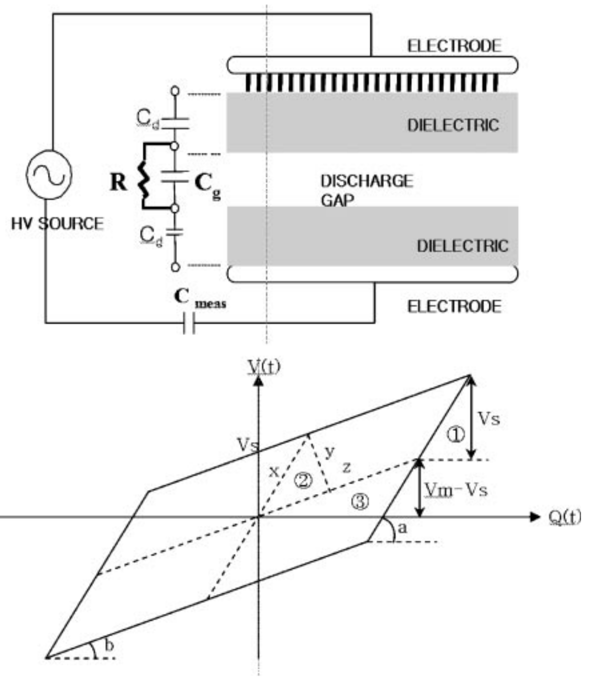


Fig. 2. Electric circuit and Q–V Lissajou plot of the atmospheric pressure plasma system (pin-to-plate type) used in this study.

2 kW) with frequencies varying from 20–30 kHz was applied to the top electrode as the AC power.

The voltage ($v(t)$) to the top power electrode was measured using a high voltage probe (Tektonix, P-6015A) and the current ($i(t)$) to the system was measured using a current probe (Pearson Electronics, 6600), which was located between the bottom electrode and the ground. In order to observe the relationship between the voltage ($v(t)$) and charge ($Q(t)$), i.e., to obtain a Q–V Lissajou curve, as shown in Fig. 2, a capacitor C_{meas} with a 10 nF capacitance was connected between the bottom electrode and the ground during the power measurements, and, the power (P) consumed in the plasma was calculated using the following equation;

$$i(t) = \frac{d}{dt}Q(t)$$

$$P = \frac{1}{T}\int_0^T v(t)\cdot i(t)dt = \frac{1}{T}\int_0^T v(t) \cdot \frac{d}{dt}Q(t)dt = \frac{1}{T}\int_0^T v(t)dQ$$

$$\therefore P = 4fC_{\mathrm{d}}\frac{1}{1+\beta}V_{\mathrm{s}}(V_{\mathrm{m}} - V_{\mathrm{s}})$$

where, f is the frequency of the AC power, C_{d} is the capacitance of the dielectric material, β is $C_{\mathrm{g}}/C_{\mathrm{d}}$, and the definitions of remaining parameters in the equation are shown in Fig. 2.

As the gas to the system, 2.5 to 10 slm He was supplied as the dilution gas to prevent a transition from a glow discharge to micro-discharge, and 0 to 5 slm of oxygen was added to the He as the process gas. A 2-µm-thick photoresist patterned on the sodalime glass for a thin film transistor-liquid crystal display (TFT-LCD) was used to examine the ashing properties of the photoresist. The photoresist etch rate was measured using a step profilometer (Tencor, Alpha Step 500). The density of the oxygen atoms in the plasma was measured using optical emission spectroscopy (OES, SC Tech., PCM-402). Field emission scanning electron microscopy (FE-SEM, Hitachi, S-4700) was used to observe the remaining photoresist residue as well as the possible physical damage to the Al–Nd metal lines on the real TFT-LCD panel after the photoresist ashing using the atmospheric pressure plasma.

3. Results and Discussion

Figure 3 shows the effect of the He flow rates at a 3 slm of oxygen flow rate and the effect of the oxygen flow rates at a 10 slm of He flow rate on the discharge current for the pin-to-plate type and the conventional DBD-type atmospheric pressure plasmas. The AC voltage was maintained at 10 kV and the air gap between the two dielectrics was maintained at 4 mm. As shown in the figure, in the case of the pin-to-plate type, the increase in the He flow rate from 0 to 10 slm increased the discharge current. Indeed, when the He flow rate was lower than approximately 2 slm, micro-discharges were observed and a stable glow discharges were observed as the He flow rate was increased. The increase in the

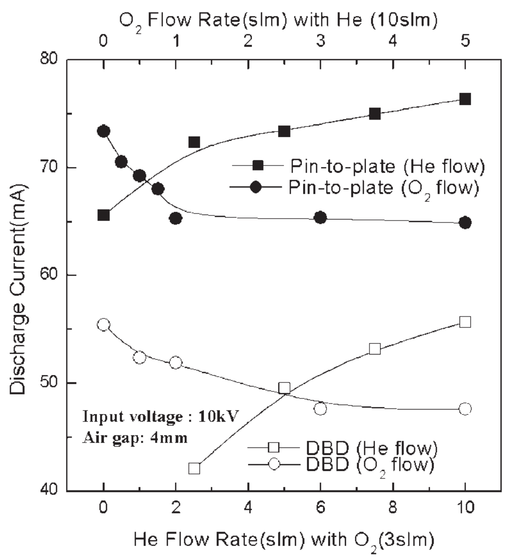


Fig. 3. Discharge current measured as a function of the He flow rate at 3 slm of O_2 and as a function of the O_2 flow rate at 10 slm of He for the pin-to-plate type and the conventional DBD-type. The input AC voltage and the air gap between the two dielectrics were maintained at 10 kV and 4 mm, respectively.

discharge current with increasing He flow rate appears to be due to the formation of a stable glow discharge over the entire electrode surface, which appears to be related to the increase in the plasma density. The increase in the oxygen flow rate from 0 to 5 slm with a 10 slm He flow rate generally decreased the discharge current, as shown in the Fig. 3. The decrease in the discharge current with the increase in the oxygen flow rate appears to be related to the decrease in the number of electrons in the plasma as a result of the formation of negative ions by the oxygen atoms and molecules.

In the case of the DBD-type, the discharge current with the increase in He flow rate and oxygen flow rates showed a similar trend to those measured with the pin-to-plate type. However, the discharge currents measured for the pin-to-plate type were approximately 1.8 times higher than those measured for the DBD type, and the discharge occurred at a lower voltage for the pin-to-plate type. The lower discharge voltage and the higher discharge current at the same voltage for the pin-to-plate type appeared to be related to the easier ionization for the pin-to-plate type. By replacing the planar electrode surface of the DBD-type with a sharp multi pin-type surface of the pin-to-plate type, a higher electric field can be obtained on the tip of the pins. Therefore, the plasma can be generated at a lower voltage. Moreover, a higher plasma density can be obtained at the same voltage than the conventional DBD-type.

Figure 4 shows the effect of the air gap distance between the dielectrics of the electrodes at an input AC voltage of 10 kV and the effect of the input AC voltage on the discharge current at the air gap of 4 mm for the pin-to-plate type discharge. The gas mixture used was He(10 slm)/O_2(3 slm). As shown in the figure, at a fixed AC voltage, the increase in the air gap from 2 to 4 mm increased the discharge current. However, a further increase in the gap to 6 mm decreased the discharge current. In the case of the AC voltage, the increase in the AC voltage from 3 to 11 kV to the multi pin-type electrode at an air gap of 4 mm increased the discharge current almost linearly. The increase in the discharge current with an increase in the air gap from 2 to 4 mm appears to be related to the increase in the plasma volume with the increase in the air gap. However, the decrease in the discharge current with an air gap >4 mm appears related to the localization of the discharge by changing the discharge from a glow discharge to a filamentary micro-discharge, as observed with the increase in the gap. The increase in the discharge current with the increase in the input AC voltage to 11 kV at a 4 mm air gap appears related to the increased ionization of the plasma without changing to a micro-discharge.

Figure 5(a) shows the etch rate of the photoresist, the relative optical emission intensity of oxygen atoms, and power consumption measured by a step profilometer, OES, and from the *Q*–*V* Lissajou plot, respectively, as a function

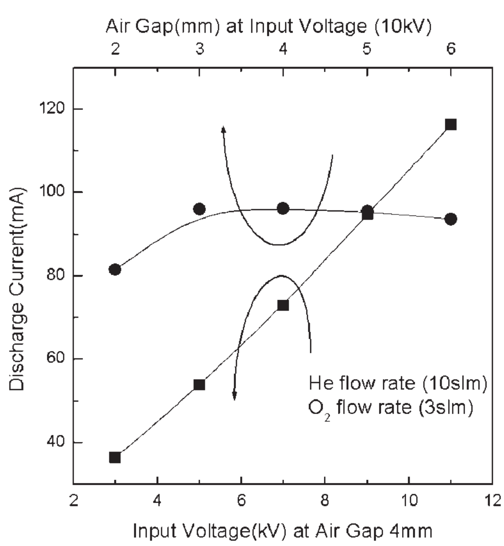
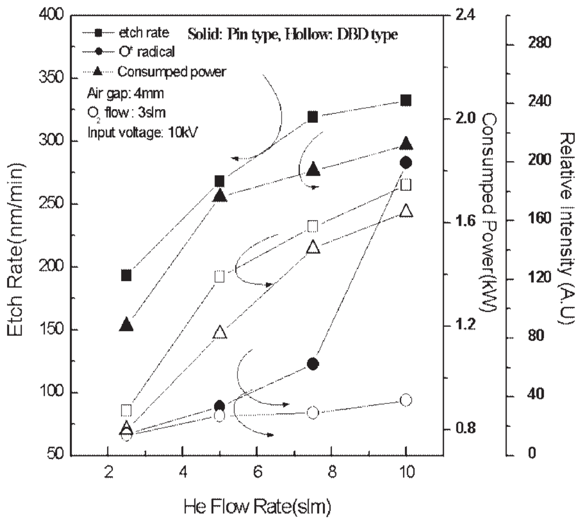
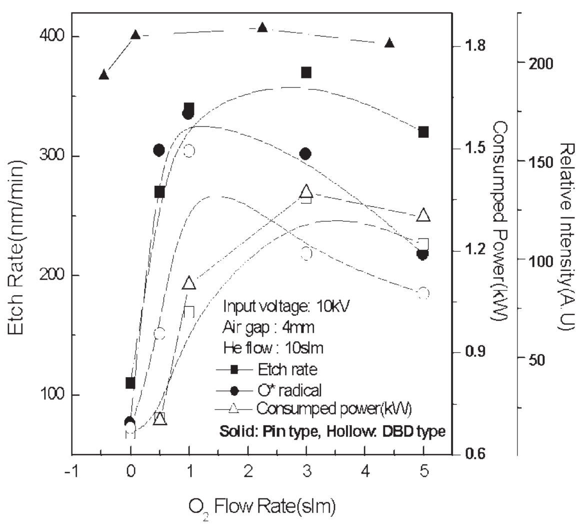


Fig. 4. Discharge current measured as a function of the input AC voltage at an air gap of 4 mm and as a function of the air gap at an input AC voltage of 10 kV for the pin-to-plate type and the conventional DBD-type. The He flow rate and O_2 flow rate were maintained at 10 slm and 3 slm, respectively.



(a)



(b)

Fig. 5. Photoresist etch rate, oxygen atomic intensity, and power consumption of the pin-to-plate type (solid) and the DBD-type (hollow) (a) as a function of He flow rate at 3 slm of O_2 flow rate and (b) as a function of O_2 flow rate at 10 slm of He flow rate for an input AC voltage of 10 kV, and 4 mm of the air gap.

of the He flow rate from 2.5 to 10 slm for the pin-to-plate type and a conventional DBD type. The oxygen flow rate, air gap distance, and the input AC voltage to the top electrode were maintained at 3 slm, 4 mm, and 10 kV, respectively. As shown in the figure, for the pin-to-plate type, an increase in the He flow rate from 2.5 to 10 slm to the system increased the photoresist etch rate from 200 to 340 nm/min. The oxygen atomic intensity measured by OES and the power consumed in the plasma at a fixed input AC voltage also increased with the increase in the He flow rate. Therefore, the increase in the photoresist etch rate with the increase in the He flow rate appears to be related to the increase in the oxygen atomic density in the plasma due to more power consumption to the plasma at the fixed AC voltage. The increased power consumption with increasing He flow rate was related to the lower breakdown voltage and the increase in the plasma-on time in the *Q–V* Lissajou plot (not shown). This is due to the increased proportion of He in the He/O_2 gas mixture with increasing He flow rate because oxygen in the gas mixture decreases the plasma density by consuming electrons in the plasma. The power can also be consumed by the dissociation of oxygen molecules by transferring the energy of the excited He to the oxygen molecules. This suggests that an increase in the He flow rate will increase the density of the excited He atoms having 19.7 eV[14] and the collision of excited He atom to oxygen molecules will easily dissociate the oxygen molecule to atomic oxygen because the binding energy of the oxygen molecule is 5.73 eV.[15] It is believed that penning dissociate ionization of oxygen molecules by excited He ($He^* + O_2 \rightarrow O + O^+ + He$; 19.3 eV) can also occur. Therefore, the increase in the oxygen emission intensity (777.5 nm) observed by OES with the increase in the He flow rate at an oxygen flow rate of 3 slm appears to be related not only to the increased plasma density but also to the increased number of oxygen atoms in the plasma as a result of the increased dissociation of oxygen molecules even though the percentage of oxygen in the plasma decreased with increasing He flow rate. Therefore, the increased photoresist etch rate with increasing He flow rate appears to be related to the increased number of oxygen atoms in the plasma.

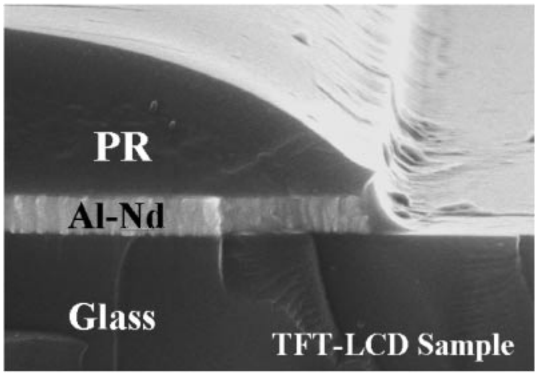
In the case of the DBD-type, the photoresist etch rate, the optical emission intensity of oxygen atoms, and the consumed power increased with the increase in the He flow rate from 2.5 to 10 slm, which is similar to the case of the pin-to-plate type. The difference was a lower photoresist etch rate, a lower OES intensitie, and a lower power consumption than those by the pin-to-plate type at the same AC voltage. Due to the low consumption of the power at a fixed AC voltage, the plasma density was lower and the oxygen atom density was lower. Therefore, the photoresist etch rates was lower than those produced by the pin-to-plate type. The measured maximum photoresist etch rate of the DBD-type was approximately 260 nm/min at He(10 slm)/O_2(3 slm) compared with 340 nm/min for the pin-to-plate type at the same gas mixture.

Figure 5(b) shows the photoresist etch rate, the relative optical emission intensity of oxygen atoms, and power consumption as a function of the oxygen flow rate from 0 to 5 slm for the pin-to-plate type and the conventional DBD type. The He flow rate, air gap distance, and the input AC voltage to the top electrode were maintained at 10 slm, 4 mm, and 10 kV, respectively. As shown in the figure, the photoresist etch rate showed a maximum at an oxygen flow rate of 3 slm for both the pin-to-plate type and the DBD-type. However, the pin-to-plate showed the higher photoresist etch rates compared with the DBD-type, which is similar to the case in Fig. 5(a). The calculated power consumption also showed a similar trend to the photoresist etch rates. In the case of the oxygen atomic intensity measured by OES, it showed a maximum at an oxygen flow rate of 1 slm, and further increases in the oxygen flow rate decreased the oxygen atomic intensity. The small addition of oxygen to He increases the photoresist etch rate by increasing the oxygen atom density in the plasma. However, the addition of oxygen decreases the plasma density due to the formation of negative ions. Therefore, the decrease in the photoresist etch rate by an oxygen flow rate >3 slm appears related to the decrease in the number of oxygen atoms in the plasma, as shown in the figure. Indeed, when the oxygen flow rate was increased from 1 to 3 slm, the oxygen atomic intensity was decreased while the photoresist etch rate and the power consumption were increased. This might be related to the formation of ozone as a result of the recombination of oxygen atoms with oxygen molecules due to the increased percentage of oxygen in the gas mixture. Therefore, even though the oxygen atom density was lower, the photoresist etch rate could be increased as a result of an increased ozone level. However, the exact reason for the increase in the photoresist etch rate with the decrease in the oxygen atomic intensity requires further investigation.

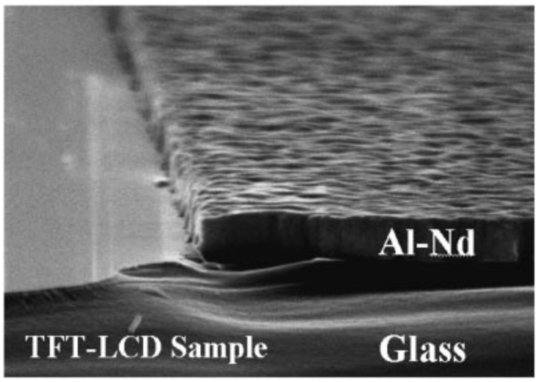
As shown in the above figures, for all of the conditions investigated at a fixed AC voltage, the pin-to-plate type showed a higher plasma density and higher processing rates than the conventional DBD-type. Therefore, the photoresist etching of a TFT-LCD substrate was performed using the pin-to-plate type. Figure 6 shows SEM micrographs of a TFT-LCD substrate (Mo/Al–Nd/glass) (a) before and (b) after the photoresist etching using the pin-to-plate type atmospheric pressure plasma. The etching condition was a gas mixture of He(10 slm)/O_2(3 slm), an air gap distance of 4 mm, and an input AC voltage of 10 kV to have a 340 nm/min of photoresist etch rate. Under these conditions, a bright glow discharge was observed over a plasma source area of 500 mm $\times$ 50 mm, and no physical damage was observed, as shown in Fig. 6(b).

4. Conclusions

In order to obtain a higher plasma density than that obtained using the conventional DBD discharge, this study used a modified DBD discharge, which has a multi-pin electrode instead of a blank plate electrode, and its discharge characteristics and photoresist etch rates were compared with those produced by the conventional DBD-type discharge. The pin-to-plate type showed a lower breakdown voltage, a longer plasma-on time, a higher discharge current, and a higher power consumption at a given AC voltage with various He/O_2 mixtures. Therefore, the pin-to-plate type appeared to be more efficient and have a higher plasma density than the conventional DBD-type. In addition, at a given voltage, the photoresist etch rate was higher for the pin-to-plate due to the higher rate of oxygen dissociation.



(a)



(b)

Fig. 6. SEM micrographs of a TFT-LCD substrate (Mo/Al–Nd/glass) (a) before and (b) after the photoresist ashing using the pin-to-plate type. The photoresist etch rate was 340 nm/min at an input AC voltage of 10 kV, He/O_2 flow rates of 10 slm/3 slm, and air gap distance of 4 mm.

When the He and oxygen flow rates were varied, there was an optimum He and oxygen mixture that showed the highest photoresist etch rate by maximizing the number of oxygen atoms and/or ozone in the plasma. Using a mixture of 3 slm of O_2 and 10 slm of He, a maximum photoresist ash rate of 340 nm/min could be obtained with the pin-to-plate type at 10 kV of AC voltage and a 4 mm air gap. When the photoresist ashing was performed with a TFT-LCD substrate, no physical damage was observed on the metal lines of the TFT-LCD devices when the above conditions were used. Therefore, it is believed that the pin-to-plate type discharge could be applied to the next generation of FPD processing.

Acknowledgments

This work was supported by the National Research Laboratory Program by Korea Ministry of Science and Technology (MOST).

1) J. Schmitt, M. Elyaakoubi and L. Sansonnens: Plasma Sources Sci. Technol. **11** (2002) A206.
2) Y. H. Lee, C. H. Yi, M. J. Chung and G. Y. Yeom: Surf. Coat. Technol. **146** (2001) 474.
3) C. H. Yi, Y. H. Lee and G. Y. Yeom: Surf. Coat. Technol. **171** (2003) 237.
4) S. Kanazawa, M. Kogoma, T. Moriwaki and S. Okazaki: J. Phys. D **21** (1988) 838.
5) S. Y. Moon, W. Choe and B. K. Kang: Appl. Phys. Lett. **84** (2004) 188.
6) H. E. Wagner, R. Brandenburg, K. V. Kozlov, A. Sonnenfeld, P. Michel and J. F. Behnke: Vacuum **71** (2003) 417.
7) Y. B. Golubovskii, V. A. Maiorov, J. Behnke and J. F. Behnke: J. Phys. D **35** (2002) 751.
8) F. Massines, P. Segur, N. Gherardi, C. Khamphan and A. Richard: Surf. Coat. Technol. **174** (2003) 8.
9) M. Simor, J. Rahel, P. Vojtek and M. Cernak: Appl. Phys. Lett. **81** (2002) 2716.
10) N. Gherardi, G. Gouda, E. Gat, A. Ricard and F. Massines: Plasma Sources Sci. Technol. **9** (2000) 340.
11) N. Gherardi and F. Massines: IEEE Trans. Plasma Sci. **29** (2001) 536.
12) F. Massines, A. Rabehi, P. Decomps, R. B. Gadri, P. Segur and C. Mayoux: J. Appl. Phys. **83** (1998) 2950.
13) K. Takaki, D. Taguchi and T. Fujiwara: Appl. Phys. Lett. **78** (2001) 2646.
14) S. C. Brown: *Basic Data of Plasma Physics* (American Vacuum Society Classics, USA, 1994).
15) Y. Yourdshahyan, B. Razaznejad and B. I. Lundqvist: Solid State Commun. **117** (2001) 531.