Evaluation of Dust Particle Properties and Particulate Contamination in a PECVD Reactor by Visualization Measurements

On-line Number 474
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ABSTRACT

The properties and behavior of dust particles generated in the plasma reactor and their effects on film contamination are studied. The motion and spatial distribution of dust particles suspended in a PECVD reactor during fabrication of a SiO₂ thin film using tetraethylorthosilicate/oxygen/nitrogen plasma are visualized by using a laser light scattering technique. It is found that the particles are in many cases trapped at a certain region of plasma. The size of the dust particles and the structure of the particle clouds trapped are strongly dependent on the plasma operating conditions, and these also in turn influence the level of particulate contamination of the substrate. It was concluded that proper process conditions must be sought carefully to achieve films of good quality and sufficiently high growth rate with minimum particulate contamination.

KEYWORDS
plasma enhanced chemical vapor deposition, dusty plasma, particulate contamination, film growth rate

INTRODUCTION

The deposition of SiO₂ films using tetraethylorthosilicate (TEOS) as a source gas in plasma-enhanced chemical vapor deposition (PECVD) has become a promising technique in very large scale integration (VLSI) technology (Tochitani et al., 1993). The SiO₂ films prepared using this technique may be used as passivation layers for interlayer isolation and lithographic masks and may be attractive as the gate dielectrics for thin film transistor (TFT) applications. This is because they exhibit good step coverage, as well as good electrical and mechanical properties (Ray et al., 1996). However, because of the low deposition rate of SiO₂ films obtained using TEOS plasma, many studies have been devoted to increase the deposition rate to meet the manufacturing requirement (DeCrosta et al., 1996). Some efforts have been undertaken for that purpose that includes increasing precursor concentration, total pressure and rf power (Raupp et al., 1992; Yi et al., 2001). Unfortunately, the approaches seem to lead to the degradation of film properties and the generation of dust particles in the reactor (Fujimoto et al., 2000; Setyawan et al., 2003).

It is well established that dust particles in the plasma are negatively charged and confined in the plasma-sheath boundaries near the electrodes that prevent them from incorporating into the growing film. Therefore, the presence of dust particles in the plasma reactor is not bad for the film properties, as long as they are not incorporated into the growing layer. However, it has been shown that dust particles may deposit into the growing films during the continuous plasma operation, not at the end of the film preparation when the plasma is turned off. Thus, it appears that dust particles can escape the plasma and cause the degradation of film properties. Hence, to find a way to control particle incorporation into films, and at the same time to obtain high deposition rate are the primary motivation of this study.

In the present work, we present the experimental investigation of particle formation and behavior in TEOS/O₂ plasma. The effects of particle behavior on film contamination are then demonstrated.
addition, the conditions under which minimum particle generation and high film deposition rate are achieved simultaneously.

EXPERIMENTAL

Figure 1 shows the experimental setup of PECVD. The PECVD reactor has been described previously (Fujimoto et al., 2000). The plasma reactor is a conventional parallel-plate type with an rf discharge of 13.56 MHz. The upper plate is in a showerhead configuration, and is coupled to the rf power supply. The lower plate is grounded. The grounded electrode contained an electrical heater equipped with a temperature controller. The electrodes are 200 mm in diameter and are separated by a distance of 35 mm. The output power of the rf generator was varied from 100 to 400 W in the forward direction, whereas the reflected power was matched to approximately zero using a matching controller.

A mixture of TEOS diluted in nitrogen gas as the carrier and oxygen were introduced into the plasma reactor through the showerhead. The ratio of oxygen to nitrogen, and the TEOS concentration were fixed at 1 and 5.0 % respectively. The flow rate was controlled by a mass flow controller (SF-2600, Tylan General) for nitrogen and oxygen and by a liquid mass flow controller (LV210, STEC, Inc.) for TEOS. The reactor was evacuated to a base pressure of less than 1.33 Pa using a dry pump (A-30, Ebara Co., Ltd.) before each experimental run.

Particle generation was detected by a laser light scattering (LLS) technique. A laser light, formed into a thin sheet using an arrangement of a rod and a cylindrical lens, was used to illuminate the space between the electrodes. The light scattered by the generated particles was then detected by a high-resolution CCD camera. In addition to the LLS technique, particle generation was monitored using a particle counter. Using this technique, a sampling probe was inserted into the reactor and the generated particles were drawn by means of a vacuum pump. The flow rate of the sucking gas was controlled in such a way that the drag by the gas flow can overcome the potential barrier for the negatively charged particles due to the floating potential of the sampling probe. The particles were then passed through a measuring cell to measure the particle number.

For the study of particle generation and behavior, the temperature could be set either ambient or 300 °C whereas for film deposition study, it was set constant at 300 °C. Silicon dioxide thin film was grown on a silicon wafer placed on the lower grounded electrode. The total gas flow rate was varied from 100 to 1000 sccm (standard cubic centimeters per minute). Film thickness was measured using a surface texture-measuring instrument (Surfcom 1400D, Accretech). The time-averaged deposition rates were determined by dividing the average film thickness by deposition time. Particles deposited on the film surface and embedded into the growing layer were observed using scanning electron microscopy (SEM).
For the case of particles embedded into the growing layers, the observations were performed on the cross sectional view of the films.

RESULTS AND DISCUSSION

Particle trapping behavior

Particle generation in the gas phase are observed in TEOS/O₂ PECVD for all conditions under study at a pressure of 4 Torr. For some conditions, particularly at high gas flow rates, the LLS technique cannot detect particles because of a very low particle concentration or a very small particle size. However, using the sampling technique described earlier, SEM observations show the presence of particles in the plasma reactor. The generated particles are trapped in the plasma/sheath boundaries near both the powered and the grounded electrodes. It appears that particle generation mainly takes place in the plasma/sheath boundaries near the electrodes as shown by the spatial distribution of the scattered light intensity in Figure 2. Peaks of the scattered light intensity appear near both electrodes. When the plasma was turned on, the system was free of particles. The particles came into sight near the electrodes immediately after the plasma was turned on. Both particle size and concentration grew over time, thereby scattering more light and making the clouds appear to become thicker. The final shape of the particle clouds is dependent on plasma operating conditions.

Figure 3 shows the dependence of the structure of particle clouds below the showerhead on total gas flow rate. The clouds are located in discrete, localized regions between the showerhead holes, and light-scattering-free regions are present just below the holes, and their surrounding areas at a certain radius, regardless of the gas flow rate. The trap regions are static points where particles are not affected so much by the gas drag force. When the gas flow rate exceeds 400 sccm, the particle clouds below the
showerhead cannot be detected by the LLS technique. For the case of a low gas flow rate, the particles form a lump cloud which is different from those observed for a high gas flow rate where the clouds are in the shape of a line. Similar phenomena have been observed previously by injecting particles from the outside that we referred to as the lumping mode and the winding mode for the cases of low and high gas flow rate, respectively (Setyawan et al., 2002b). When the gas flow rate is high, the neutral drag force presses the clouds more strongly without changing the equilibrium position of the trap that causes the clouds to shrink. It appears that the origin of the particles, i.e., whether they are injected from the outside or are generated somewhere inside the plasma reactor, has no effect on the structure and trap location of the particle clouds below the powered electrode.

When the rf power is increased, the particle clouds become smaller and thinner. Upon increasing the rf power, the ion density increases, thus increasing the ion drag force, since this force is proportional to the ion density (Barnes et al., 1992). The ion drag force drives the particles toward the showerhead surface. Moreover, the electric field in the sheath becomes stronger with increasing rf power (Edelberg and Aydil, 1999). The electrostatic force acting on the charged particles then becomes stronger because this force is proportional to the intensity of the electric field. The electrostatic force drives the particles away from the showerhead surface. The two stronger forces, i.e., the ion drag and electrostatic, that act oppositely on the particles compress the particle clouds and they become smaller and thinner.

The particle clouds at a substrate temperature of 300 °C were more compressed compared to the particle clouds at room temperature. In addition, the clouds were closer to the powered electrode. When the substrate is heated to 300 °C, a temperature gradient exists between the heated grounded electrode and the powered electrode that is not heated. Therefore, the particles are pushed toward the powered electrode by thermophoretic force that is present due to the temperature gradient. This causes the clouds to become more compressed and to be located closer to the electrode.

Particle formation and growth

The visualizations described above only provided information concerning the structure and spatial distribution of particles in the trap. In order to understand particle formation and growth, the intensity of the scattered light in the trap regions near the electrodes was examined. It is not possible to obtain separate information on both size and particle concentration from the scattered intensity, but it provides information on the combination of both. The particle size was deduced from scanning electron microscopy (SEM) images of particles deposited on the Si wafer by drawing the particles from the trap using the sampling probe described earlier.

Figure 4 shows the effect of gas flow rate on particle formation at room temperature. A similar trend can be observed for the condition where the substrate temperature is 300 °C. The scattered light intensity decreases when the total gas flow rate is increased. It appears that particle growth is suppressed at high gas flow rates. The residence times of TEOS vapor, intermediates, and primary particles in the plasma reactor become longer when the gas flow rate is decreased. This provides more time for the primary particles formed to grow,
resulting in a larger particle size (Kim and Ikegawa, 1999). When the growing particles reach a critical size or concentration, above which the plasma cannot confine them any longer, they are released from the trap. The particles can also be observed to escape from the trap through the edge of the powered electrode. Because of the low residence time of the process gases, a high gas flow tends to remove potential particle generation from the sheath before they are able to form particles in the nucleation sites and provides the less opportunity for the primary particles to grow. SEM photographs of the particles show that the particles are larger at a low gas flow rate and are smaller at a high gas flow rate. This will be discussed below.

Figure 5 shows examples of SEM images of particles drawn and deposited onto the Si wafer using the sampling technique described above for the conditions at room temperature and a substrate temperature of 300 °C. The total gas flow rate is 60 sccm. The particles were drawn from the trap when the plasma duration reached 30 sec. The particles are composed of smaller primary particles that serve as precursors for larger agglomerated particles. This suggests that the particles in the TEOS/O₂ plasma are grown by coagulation. The primary particles are spherical in shape. For the case of the substrate temperature of 300 °C, the boundary among the primary particles is not clear, suggesting that surface deposition has occurred.

**Effect of particle trapping behavior on particle contamination**

In order to clarify the effects of particle trapping behavior on particle contamination on the silicon wafer, a silicon wafer was placed on the grounded electrode. It was then exposed to the TEOS/O₂ plasma for about 5 min. Particle deposited on the wafer were observed using SEM. We observed that the plasma duration is sufficient to allow the formation of stable clouds below the powered electrode. Figure 6 shows SEM images of particles deposited on the wafer for gas flow rates of 60 sccm, 200 sccm, and 300 sccm, respectively, at room temperature. Numerous particles were deposited on the wafer for gas flow rates of 60 sccm and 300 sccm and only a few for a gas flow rate of 200 sccm. It appears that particle deposition decreases with increasing gas flow rates, then reaches a minimum level and eventually increases again. The particles deposited on the wafer placed on the grounded electrode have the sizes in the range of particle size observed in the trap for each gas flow rate. This suggests that they originate from the trap regions and are deposited on the wafer after the plasma is turned off, as was also observed in the previous study (Setyawan et al., 2003). Because the particles fall on the wafer after the plasma is turned off, the level of particle contamination on the wafer will be influenced by the size and concentration of particles in the trap. At low gas flow rates, the generated particles trapped near the powered electrode are large,
and they tend to deposit on the wafer because they cannot follow the gas pathlines when they approach the wafer. At higher gas flow rate, more particles are trapped near the grounded electrode.

Effect of gas flow rate on film growth

Films deposited using TEOS/O₂ plasma at varying gas flow rate and at a rf power of 100 W and a pressure of 2 Torr provided growth rate ranging from 0.12 to 0.28 µm/min, as shown in Figure 7. The film nonuniformities, represented by coefficient of variation that is defined as standard deviation/mean, are in the 3-10% range. The film growth rate increases with increasing gas flow rate, reaches a maximum value, and then decreases again. On the other hand, the film nonuniformity is relatively constant at low gas flow rate (<400sccm) and then increases monotonously with increasing gas flow rate. It appears that at high gas flow rate the deposition rate is low and the uniformity is poor.

Figure 8 shows SEM images of particles deposited on the silicon wafer placed on the powered electrode at a gas flow rate of 100 sccm. The particles are agglomerate, suggesting that they grew through coagulation. The shape of the agglomerated particles suggests that they were formed by the fusion of primary particles as also observed previously by Fujimoto et al. (2000). This indicates that the primary particles are soft or melting, like a gel or liquid that is probably caused by the physical properties of TEOS, since this shape was also observed in TEOS/O₂ atmospheric pressure chemical vapor deposition (APCVD) process (Adachi et al., 1992). It seems that particle deposition takes place after the plasma is turned off. The particles are attracted to the powered electrode once the rf power is turned off due to the drag of ion flow towards the electrode that retains its negative self-bias voltage. The number of particles deposited on the powered electrode becomes fewer with increasing gas flow rate. This is related to the effect of gas flow rate on particle formation and trapping in the sheath region near the powered electrode. High gas flow rate tends to suppress particle formation as shown in Figure 9 in which the intensity of the light scattered by
Particles in that region decreases with increasing gas flow rate. Moreover, high gas flow rate provides large gas drag that will carry the particles away from the trap region near the powered electrode. More detail discussion about this effect has been given previously (Setyawan et al., 2004).

We observed previously that dome-like structures were formed on the film surface due to the deposition of submicrometer particles during continuous plasma operation that could not be leveled off easily by film deposition. In order to confirm this, we observed the cross sectional view of the films grown on the silicon wafer placed on the grounded electrode using SEM. Figure 10 shows the SEM photographs. At a low gas flow rate, i.e. 100 sccm, no particles are present in the film, whereas at a high gas flow rate, i.e. 1000 sccm, particles are observed to be embedded into the growing film. Thus, it can be proven that the particles are incorporated into the growing film during the deposition process. The size of the embedded particles is about 150 nm, which is in the range of particle sizes observed in TEOS/O₂ plasma space. Even though dust particles of submicrometer size are confined in the plasma, some fractions of the dust particles are considered to escape from the confinement to cause deposition under certain plasma conditions.

![Figure 8. SEM images of particle deposited on the silicon wafer placed on the powered electrode at a gas flow rate of 100 sccm.](image)

![Figure 9. Dependence of particle formation on gas flow rate.](image)

![Figure 10. SEM images of cross sectional view of silicon dioxide thin film showing the effect of gas flow rate on particles embedded into the growing film: (a) no embedded particle at 100 sccm, and (b) particles are embedded into the film at 1000 sccm.](image)
CONCLUSIONS

Particle formation, trap and contamination in TEOS/O\textsubscript{2} plasma have been studied by an in situ laser light scattering technique and ex situ scanning electron microscopy. The film growth rate and uniformity were also measured. The particle clouds are dependent on plasma parameters such as rf power, gas flow rate, and substrate temperature. The particles form a lump shape at low gas flow rates, change to a line shape with increasing gas flow rates, and finally the LLS technique is not able to detect them any longer at high gas flow rates. When the rf power is increased, the particle clouds are compressed by the stronger electrostatic and ion drag forces acting oppositely on the particles. By heating the grounded electrode to 300 °C, thermophoresis due to the temperature gradient shifts the particle clouds upward towards the powered electrode.

The particle formation and growth is suppressed with increasing gas flow rate. The deposition rate of silicon dioxide thin film increases with increasing gas flow rate, reaches a maximum value and eventually decreases again. However, the uniformity of the film tends to degrade at high gas flow rate. At high gas flow rate, some radicals responsible for particle generation are carried away from the nucleation site near the showerhead-type powered electrode to the sheath region near the substrate. Then, particles are generated there and at a particular condition, some of them pass through the sheath to reach the substrate and are embedded into the growing film.

ACKNOWLEDGEMENTS

This work has been supported by Innovation Plaza Hiroshima of JST (Japan Science and Technology Agency) and a Grant-in-Aid for Scientific Research by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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