Role of hydrogen in evolution of plasma parameters and dust growth in capacitively coupled dusty plasmas

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The temporal behavior of naturally produced dust parameters (radius and density) and plasma parameters (electron temperature and ion flux) was investigated in radio frequency SiH₄/H₂/Ar plasmas. As a result, the electron temperature and ion flux were shown to be strongly correlated with the three-step dust growth pattern. In addition, the generation of dust particles was suppressed by mixing more hydrogen gas due to the plasma chemistry, and consequently, the dust growth rate in the molecular accretion growth, which is known to be proportional to the growth rate of thin film deposition, increased.


Dusty plasmas are ionized gases containing tiny solid particles. They are ubiquitous in space and laboratory plasmas such as interstellar clouds, Saturn’s rings, industrial processing plasmas, and tokamak plasmas. Among them, SiH₄/H₂ plasmas, which are actively used for growing Si films, have been of great interest in the photovoltaic and semiconductor manufacturing. In this field, the methods to obtain high film growth rates and crystalline films have been intensively studied. However, since Si based dust particles are generated spontaneously in SiH₄/H₂ plasmas, significant changes in the plasma parameters and deterioration in product reliability have been reported. Moreover, these issues are becoming more significant as the area of processing plasma grows larger and the feature size of semiconductors decreases. Therefore, it is inevitable to investigate how dust particle generation and growth affect SiH₄/H₂ plasmas.

Electron temperature, known as the average kinetic energy of electrons, is one of the most important parameters because electrons govern ionization, excitation, and dissociation processes in the plasma. However, since dust particles are spontaneously produced in dusty plasma, accurate measurement of various plasma parameters is generally not straightforward. For instance, although Langmuir probes are widely employed in low temperature plasmas, it is impossible to collect an accurate plasma current with a conventional Langmuir probe due to probe tip contamination caused by chemical reactions occurring inside the plasma. Instead, it is possible to measure relative values of plasma parameters by the plasma emission spectroscopy. Therefore, in this work, we adopted a recently developed floating probe to measure the plasma parameters.

The goals of this work are to investigate the behavior of the electron temperature and ion flux with particle growth and to study the hydrogen effect on the plasma and dust behavior. To achieve these, the dust parameters and plasma parameters were measured in various hydrogen partial pressures. In addition, the role of hydrogen in SiH₄/H₂/Ar plasmas with dust particle growth is discussed.

Figure 1 shows a top view of the experimental setup. The plasma source is a capacitively coupled type, and the electrode is powered by a 13.56 MHz power supply (RFPP RF10S). The silane diluted argon gas (consisting of 5% SiH₄ and 95% Ar) and additional hydrogen gas were used to produce plasma. In this work, while SiH₄/Ar gas was fixed at 34 mTorr, the hydrogen partial pressure was varied from 0 to 9 mTorr to investigate the role of hydrogen in SiH₄/H₂/Ar plasmas.

A copper grid was set beside the electrode to collect dust particles. The radius of the collected dust particle was determined by transmission electron microscopy (TEM) images. The dust particle density was obtained by the laser extinction method (LEM), the details of which are found in our previous paper. In this work, the multipass LEM was adopted to lower the detection limit by installing two highly reflective spherical mirrors on both sides of the reactor chamber, as shown in Fig. 1.

The floating probe was employed to measure the electron temperature and the ion flux. In contrast to the conventional Langmuir probe, the floating probe acquires an ac current rather than a dc current by biasing a 50 kHz ac sinusoidal waveform to the probe tip. Therefore, it is only slightly influenced by the probe tip coating unless the deposited film is too thick. In the probe measurement, the probe tip was positioned vertically at 5 cm from the electrode surface to minimize perturbation.

![FIG. 1. (Color online) A schematic illustration of the experimental setup.](image-url)
A residual gas analyzer (Balzers QMS 200) was used to measure residual gas components during the discharge. Gas components such as SiH₄, Ar, and H₂ were monitored during the discharge on and off.

Figure 2(a) presents the dust radii measured by the TEM images. The squares, triangles, and circles correspond to the 0, 3, and 6 mTorr hydrogen partial pressure cases, respectively. The total gas pressure was 34, 37, and 40 mTorr, respectively. The input rf power was fixed at 50 W.

Consistent with other previous reports, our experimental result showed three different growth steps: the nucleation growth occurred right after the plasma ignition (0 s), the coagulation growth followed until around 100 s (50 s for 3 and 6 mTorr cases), and then the molecular accretion took place after that. It is noticeable that the molecular accretion growth occurs early and the dust growth rate increases in the molecular accretion phase, while it remains almost the same in both the nucleation and coagulation phases as the hydrogen partial pressure increases. At 200 s, the particle radius was measured to be 45 nm for the no hydrogen addition case. On the other hand, it was 80 nm at 6 mTorr hydrogen. It is generally known that the particle growth rate during the molecular accretion step is proportional to the growth rate of the thin film. Therefore, our experimental result suggests that the film growth rate may be improved with high hydrogen partial pressure during the molecular accretion step.

The measured dust density depicted in Fig. 2(b) clearly shows that it is smaller with higher hydrogen partial pressure. At 200 s, the dust density of the no hydrogen case is more than ten times higher than that of the 6 mTorr case. In temporal, the dust density decreases gradually.

The measured electron temperature and ion flux are described in Fig. 3, where solid, dashed, and dotted curves represent 0, 3, and 6 mTorr hydrogen cases, respectively. The electron temperature first increases and then decreases slowly in time. This is closely related to the dust growth step. During the coagulation growth step, the amount of electrons collected to dust particles rapidly increases as the dust particles become larger, and this brings about a considerable decrease of electron density (which can be less than 1/10 of ion density in the silane plasma). As a result, the electron temperature steeply increases to balance the generation against loss of electrons and ions to the dust and the wall. After the coagulation growth, the electron temperature follows the total surface area of the dust particles levitated in the plasma according to our experiment, which will be reported elsewhere. Therefore, it is observed that the electron temperature decreases slowly in time during the molecular accretion growth step.

In addition, the electron temperature is slightly lower with the lower hydrogen partial pressure during the nucleation growth step (0–5 s). It is believed that the electron temperature is lowered by incoming neutral hydrogen molecules, which was also observed in the Ar/H₂ plasma. Furthermore, while the measured electron temperature is almost the same under the various hydrogen partial pressures during 5–50 s, it behaves differently during 50–200 s. In detail, the electron temperature starts to decrease after 100 s in case of no hydrogen, but it decreases from around 50 s for 3 and 6 mTorr hydrogen cases. This indicates the starting point of the molecular accretion step.

In the ion flux measurement, two peaks exist, the first of which appears at 5 s. The second peak shows up at 50 s for 3 and 6 mTorr hydrogen cases and at 75 s with no hydrogen addition. It can be seen that the second peak of the ion flux indicates the end of the coagulation growth. It is noteworthy that the starting point of the molecular accretion growth is shifted earlier by injecting hydrogen gas.

The temporal behavior of the ion flux is related to the electrical power dissipated in the plasma. Because the plasma impedance changes in accordance with dust growth, the dissipated power to the plasma also varies in time. In addition, after the coagulation growth, dust particles become major charge carriers rather than electrons because of the sharp drop of electron density during the coagulation phase due to the large electron flow to the dust. Therefore, the plasma becomes more resistive and the electrical power absorbed by plasma increases, which causes the increase of the ion flux.

At the same time, SiH₄ consumption was investigated by the residual gas analyzer, and the result at 100 s is shown in Fig. 4. The SiH₄ consumption was obtained by subtracting the measured SiH₄/Ar intensity during the discharge off time.
from that during the discharge on. Note that the consumption of SiH₄ is reduced at higher hydrogen partial pressure. This suggests that the amount of generated dust particles is reduced with the raised hydrogen partial pressure. The result is consistent with other previous reports. The reason for this is related to plasma chemistry. Disilanes such as Si₂H₄ and Si₂H₆, produced from chemical reactions among SiH₂, SiH₃, and SiH₄, play a critical role in the dust generation. However, since the disilane dissociation rate to monosilanes is proportional to the hydrogen molecule density, the dust generation is suppressed by adding more hydrogen gas.

In addition, the increased growth rate in the molecular accretion step is explained as follows. As the dust generation decreases, the electron loss to dusts decreases, and as a result, the electron density increases. Therefore, SiH₃ molecules, which are produced by the reaction between electrons and SiH₄ molecules, are created in greater numbers. This leads to the increase of the growth rate during the accretion step.

In summary, we measured the temporal behavior of spontaneously generated dust size and plasma parameters in capacitively coupled SiH₄/H₂/Ar plasmas. As a result, the electron temperature increases during the coagulation growth and slowly decreases thereafter. In addition, the ion flux shows two maximum values at the nucleation and at the end of coagulation growth. Moreover, the amount of generated dust particles is reduced when more hydrogen gas is injected because the production of higher-silane, the seed of dust particles, is hindered by hydrogen molecules. Thus, the electron density in the molecular accretion growth step is high, and therefore the dust growth rate increases at high hydrogen partial pressure.

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