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Detection of particulates in a rf plasma by laser evaporation and subsequent discharge formation

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Nd:YAG-laser-induced evaporation of particulates formed in an Ar-CCl$_2$F$_2$ rf plasma and the subsequent discharge in the vapor have been investigated in situ by means of optical emission spectroscopy. The estimated threshold for discharge formation is $5 \times 10^6$ W/cm$^2$. The maximum laser-induced emission intensity is observed when the laser is operated in the long-pulse mode (about 200 µs pulse duration) at the fundamental frequency. The wavelength integrated intensity of this continuum emission has been compared with light scattering intensity at the same laser energy. It has been found that the laser-induced emission intensity can be more than ten times higher than the scattering intensity, especially for particulates with a diameter much smaller than the wavelength of the laser. Therefore, this effect provides a sensitive particulate detection method.

It is generally accepted that particulates formed in a rf plasma cause major damage in surface modification processes such as deposition, sputtering, and etching. The main problem in the investigation of particulates in these plasmas is the lack of an efficient and sensitive diagnostic, which would allow determination of particulate size and density in situ and spatially resolved. A low detection limit of such a technique is essential not only for fundamental research but also for the control of the contamination level in industrial surface processing. The commonly used diagnostic is light scattering. This method is ideal for a qualitative study, but major problems can be encountered in obtaining absolute figures. A quantitative analysis in the Mie scattering regime (i.e., when the wavelength of the light is in the order of or smaller than the particulate size) requires knowledge of size and shape distribution and refractive index of the particulates, and besides it is computationally complex. Moreover, the complete angular distribution and state of polarization of the scattered light intensity should be measured. For smaller particulates in the Rayleigh regime stray light determines the detection limit due to a very low scattering intensity. Therefore, an effort has been made to provide an alternative method which eliminates the problems of light scattering.

Here we present a detection technique based on laser evaporation of particulates. This method uses the fact that the particulates absorb laser light and consequently evaporate into a dense cloud. Further deposition of laser power in the cloud leads to a laser-induced breakdown. This phenomenon is visible as a white flash in the plasma. The possible mechanisms leading to breakdown in the vapor are inverse bremsstrahlung (IB), dominant at high pressures and/or electron densities, and multiphoton ionization (MPI). The gas density in a rf plasma is itself too low to cause any kind of breakdown at the used laser power. This has been experimentally verified in our case.

In this work the particulates are produced in a rf plasma, operated at 13.56 MHz. A parallel-plate configuration, with an electrode diameter of 12 cm and an inter-electrode distance of 5.5 cm, is used. On top of the water-cooled lower-rf-powered electrode a silicon wafer can be placed. The feed gas is introduced homogeneously through a circular slit around this electrode. The evaporation and scattering experiments are performed using a Nd:YAG laser. It can be operated in a Q-switched or a long-pulse mode, resulting in one short pulse of about 5 ns or a train of pulses spread over 200 µs. The maximum energy per pulse is 0.45 J at the fundamental frequency ($\lambda = 1064$ nm) and 0.35 J at the doubled frequency. The donut-shaped laser beam is collimated into a diameter of 2–3 mm. The detection system consists of an EG&G OMA III system, supplied with a 0.25 m Jarrel-Ash monochromator with a 150 or 1200 grooves/mm grating and a 0.35 J at the doubled frequency. The laser-induced emission intensity is observed when the laser is operated in the long-pulse mode (about 200 µs pulse duration) at the fundamental frequency. The wavelength integrated intensity of this continuum emission has been compared with light scattering intensity at the same laser energy. It has been found that the laser-induced emission intensity can be more than ten times higher than the scattering intensity, especially for particulates with a diameter much smaller than the wavelength of the laser. Therefore, this effect provides a sensitive particulate detection method.

Abundant particulate formation has been reported by Selwyn and Selwyn, Heldenreich, and Haller under the following conditions: 90% argon, 10% CCl$_2$F$_2$ at 200 mTorr and 100 W power input and total gas flow of 40 scm, in the presence of a Si wafer on the electrode. For these conditions we observe aggregation of particulates in the lower plasma-sheath boundary after about 5–20 min of plasma operation, dependent on the contamination of the reactor surface. The particulates are confined in two separate regions: a dome above the wafer and a ring around it. These particulates can reach millimeter sizes and they have a characteristic branched chain shape. Laser evaporation experiments have been performed under the above conditions. The laser-induced emission decreases after 10–100 shots at a 10 Hz repetition rate and it finally disappears. This is due to the destructive nature of laser evaporation in combination with a low particulate formation rate. In this way we can "clean" separately the two regions in which...
scattering and stray light is 10 times higher than the continuum emission level. The plasma emission has been subtracted. The peak at 1064 nm due to induced emission exhibits a threshold behavior, which is in agreement with the breakdown character of the process. The threshold is 50 mJ/pulse, which corresponds to a laser fluence of about 10² J/cm². Furthermore, the effect starts to saturate at high laser intensities. It is evident that above threshold the continuum emission intensity is higher than the scattered intensity.

Comparing the ablation efficiency for two wavelengths in the Q-switched modes we find that the shorter wavelength (532 nm) results in more than 100 times lower emission intensity than the fundamental one (1064 nm). Since the IB absorption coefficients and breakdown thresholds scale as λ² and λ⁻⁴, respectively, the observed wavelength dependence suggests an IB-dominated process. A MPI-dominated breakdown would show an opposite dependence on the photon energy. The laser-induced emission intensity is about 10 times higher in the long-pulse mode at the same total pulse energy. This is in contrast to a "normal" laser breakdown in gases, which can be performed practically only in the Q-switched mode. It indicates that energy transport within particulates during the evaporation phase is limiting the rate of the total process. The absence of breakdown at shorter laser wavelengths does not exclude particulate evaporation; on the contrary, it occurs readily in these conditions.

It has been found that the relative sensitivity of the evaporation technique increases for smaller particulates. These are readily formed in a plasma containing only few percent of CCl₂F₂ in argon. In these conditions a laser-induced emission signal can be observed already after several seconds of plasma operation. The signal does not change as a function of the number of shots due to a much higher formation rate than in the previous case. The scattering signal is too weak to be recorded, which indicates that the diameter of particulates is below 100 nm (Rayleigh regime). The spatial distribution of the particulates has changed as well, and it becomes uniform throughout the plasma glow. The emission spectrum remains the same as that for large particulates, suggesting that the nature of the effect does not change.

From the above experiments it follows that breakdown in a dusty plasma is easy to observe due to its relatively low threshold. The integrated laser-induced emission intensity is about one order of magnitude higher than the scattered intensity in the Mie regime and probably even more for smaller particulates. In contrast with scattering, a high stray light level does not disturb the detection. Further investigations using a better time resolution have to decide on the nature of the emission, resulting in a physical model of the evaporation and excitation processes. From this a
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