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

Citation for published version (APA):

DOI:
10.1063/1.354603

Document status and date:
Published: 01/01/1993

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher’s website.
- The final author version and the galley proof are versions of the publication after peer review.
- The final published version features the final layout of the paper including the volume, issue and page numbers.

Link to publication

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the “Taverne” license above, please follow below link for the End User Agreement:
www.tue.nl/taverne

Take down policy
If you believe that this document breaches copyright please contact us at:
openaccess@tue.nl
providing details and we will investigate your claim.
Detection of particulates in a rf plasma by laser evaporation and subsequent discharge formation

W. W. Stoffels, E. Stoffels, G. M. W. Kroesen, and F. J. De Hoog
Department of Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

(Received 21 October 1992; accepted for publication 18 April 1993)

Nd:YAG-laser-induced evaporation of particulates formed in an Ar-CCl_2F_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 5×10^6 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,^{1,2} sputtering,^{3} and etching.^{4} 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).^{5} 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 (λ=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. The laser-induced plasma radiation is collected at 90° by focusing it onto a quartz fiber. The laser triggering and data acquisition timing are controlled by the OMA system, with a repetition frequency of 1 Hz and a minimum exposure time of 16 ms. The plasma emission is subtracted at the end of each cycle. An intensity calibration has been performed using a tungsten ribbon lamp.

Abundant particulate formation has been reported by Selwyn^{4} and Selwyn, Heldenreich, and Haller^{6} under the following conditions: 90% argon, 10% CCl_2F_2 at 200 mTorr and 100 W power input and total gas flow of 40 sccm, 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...
the particulates are confined, which implies that transport between these two regions is blocked.

In Fig. 1 the emission spectrum of evaporated particulates is presented. The spectrum is dominated by continuum radiation. Since the exposure time of our OMA system is relatively long the emission is integrated over the evaporation and breakdown as well as the extinction phase; therefore, the origin of the continuum is not clear. The possibilities are electron-ion or electron-neutral free free emission from the plasma or a blackbodylike emission from heated particulates. The best free-free fits (300 nm > \lambda > 1000 nm) indicate an electron temperature of about 5000 K, whereas we expect temperatures of several times 10^4 K, typical for a laser-induced breakdown. Besides, the fits predict up to two orders of magnitude more intensity in the UV region (300–450 nm) than observed. A blackbody fit yields a temperature of 3000 K, which is reasonable and the deviations in the UV region are less than a factor of 10. A better time resolution will allow to separate several phases in this process. Moreover, atomic and molecular emission, dominant in the extinction phase, will give information about the composition of the particulates. On top of the spectrum at 1064 nm the onset of the scattering signal (not corrected for the stray light) is visible. In Fig. 2 the scattering intensity is compared with the integrated laser-induced emission from 600 to 1040 nm as a function of laser energy. The laser is operated in the long-pulse mode at the fundamental frequency and fired through the plasma-sheath boundary, where the relatively large particles accumulate.

Since the IB absorption coefficients and breakdown thresholds scale as \lambda^2 and \lambda^{-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 \text{CCl}_2\text{F}_2 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
quantification of number and size of the evaporated partic-
ulates might be obtained.

The authors would like to thank Jeroen Langevoort from Philips Lighting, Eindhoven, for enabling us to use the OMA system. The work was supported by the Commission of the European Community under Brite Euram Contract No. BREU-CT91-0411.
