Electron transport coefficients in dusty argon plasmas

Michael J. McCaughey and Mark J. Kushner
University of Illinois, Department of Electrical and Computer Engineering, Gaseous Electronics Laboratory, 607 E. Healey, Champaign, Illinois 61820

(Received 1 June 1989; accepted for publication 10 July 1989)

Low-temperature partially ionized plasmas, as used in plasma processing reactors and gas lasers, are often contaminated by gas phase particulates (1–10's μm radius) resulting from electrode sputtering or gas phase chemical reactions. Particles having sizes comparable to or greater than the Debye length will negatively charge in the plasma and form a sheath at their surfaces. These particles thereby become a Coulomb-like scatterer of electrons. A hybrid Monte Carlo/molecular dynamics computer simulation has been developed to study the effect of such particulate contamination on electron transport in glow discharges and this letter presents results for argon. The dominant effect of particulate contamination is to shift the electron energy distribution to lower energies, thereby reducing electron impact rate coefficients for processes which have high threshold energies, particularly ionization. The self-sustaining E/N of discharges having dusty plasmas is therefore increased. The effect, proportional to particle density, begins to become important at a density of 10^3–10^5 cm^{-3} for gas pressures of 0.1–3 Torr.

Dust grains and gas phase particulates, or "dust," of many micron and submicron sizes have been observed in a variety of low-temperature plasma devices including lasers,[1] plasma processing reactors,[2,3] and magnetohydrodynamic (MHD) generators,[4] as well as planetary atmospheres.[5,6] Gas phase particulates in glow discharge systems are typically produced by sputtering from surfaces or from gas phase polymerization.[7–9] Many analytical studies of the effect of dust on the properties of space plasmas have been performed for conditions where the Debye length λ_D is much greater than the average distance between particles.[5,6] However, in laboratory glow discharge devices, λ_D is typically much less than the distance between particles. If the size of the particle is comparable to or larger than λ_D, the particle negatively charges and a sheath forms at its surface. Electron transport may, therefore, be perturbed near the dust due to Coulomb-like collisions. Particulates in plasma deposition systems, observed by laser scattering,[2,3] are found dominantly near the edges of the sheaths thereby suggesting a negative charge state. The presence of gas phase particulates in glow discharges used for CO_2 lasers has been linked to plasma instabilities such as streamer formation and arcing,[10] and to breakdown in pulse power devices.[11] Additionally, the presence of dust in plasma deposition systems is known to have adverse effects on the quality of the deposited film.[12]

The charging of aerosols[13] and secondary emission from particles in glow discharges[3] has been previously investigated. The effect of particulates on electron transport properties, though, is not known. In order to evaluate these effects, a computer model has been developed to calculate electron transport coefficients in "dusty" glow discharges. Using this model, electron transport in dusty argon plasmas has been examined for a variety of dust densities, dust radii, and electric field values. Results from that study will be presented in this letter.

If the size of a dust particle is commensurate to or larger than λ_D, it appears macroscopically large. If so, the surface of the dust in an electropositive plasma becomes negatively charged due to the excessive impingement of electron flux onto its surface from the plasma compared to the positive ion flux. This charging results in a non-neutral region, or sheath, developing adjacent to the surface of the dust. In analogy to the sheath which develops at large dielectric surfaces in contact with a plasma, a large dust particle in a Maxwellian plasma will acquire a potential

$$\phi_s = -(kT_e/2q)\ln(T_e M_I/T_I m_e), \qquad (1)$$

where T_e and T_I are electron and ion temperatures, and m_e and M_I are the electron and ion masses.[14] This negative potential and associated sheath result in a shielding volume several λ_D in extent around the particle. The shielding volume deflects electrons convecting under the influence of the externally applied electric field. The effect of dust on electron transport is, therefore, manifested by the perturbing effect of this deflection on the local electron energy distribution and, when averaged over the plasma, the electron impact rate coefficients.

The model we have developed to evaluate these effects is a hybrid simulation using both Monte Carlo (MC) and molecular dynamics (MD) methods. Far from a dust particle, electron transport resulting from collisions with gas atoms is calculated using conventional MC techniques in a uniform electric field. When an electron approaches a dust particle, the equations of motion are integrated with MD techniques using the electric field described below. Collisions with the neutral background gas are simultaneously accounted for at intervals given by the MC algorithms.

In a real plasma, the electron energy distribution (EED) is not necessarily Maxwellian and, therefore, Eq. (1) is not strictly valid. ϕ_s can, however, be determined by that value which balances the electron flux and ion flux to the surface of the dust. The model accounts for the non-Maxwellian nature of the plasma by calculating ϕ_s in this manner and updating the sheath potential over time to reflect changes in the electron energy distribution. ϕ_s is obtained from

$$\Gamma_e = \frac{n_e}{4}\int_{-e\phi_s}^{\infty} f(\epsilon)\left(\frac{2\epsilon}{m_e}\right)^{1/2} d\epsilon = \frac{n_I V_I}{4} = \Gamma_I, \qquad (2)$$

where Γ is the electron or ion flux, V_I is the ion thermal velocity, and $f(\epsilon)$ is the EED. This expression accounts for the fact that only electrons with energy greater than ϕ_s can reach the surface of the dust.

The electric field in the shielding volume around the dust was approximated as a $1/r$ Coulomb potential with Debye shielding

$$\phi(r) = \phi_s(d/r)\exp[-(r-d)/l], \qquad (3)$$

where d is the radius of the dust particle and r is the radial distance of the electron from the center of the particle. The characteristic radius of the shielding volume radius, l, is the minimum of λ_D and the distance required for the shielding electric field to decay to 10% of the applied electric field in half the interparticle spacing. This latter constraint is necessary only at high dust densities in order to avoid the unphysical condition of the Debye spheres of adjacent dust particles overlapping. The force due this potential is then added to the applied electric field and used in the molecular dynamics portion of the model.

The electron energy distribution in an Ar discharge calculated by the model for pristine and dusty plasmas is shown in Fig. 1. The conditions are $E/N = 7$ Td (1 Td $= 10^{-17}$ V cm^2), $P = 0.1$ Torr, and $T_{\rm gas} = 300$ K. The dust density is $\rho = 10^5$ cm^{-3} and the electron density is 5.0×10^{11} cm^{-3}. These parameters were chosen to simulate a gas laser or magnetically assisted plasma-processing reactor. The scattering of electrons from the Debye shield around the dust causes a reduction in the high-energy component of the EED and an increase in the low-energy component.

The ionization rate coefficient for pure argon in pristine and dusty plasmas ($\rho = 10^5$ cm^{-3}) as a function of applied E/N is shown in Fig. 2. The depression of the high-energy component of the EED results in a decrease in the ionization rate coefficient k_I. The difference between ionization coefficients in the pristine and dusty plasmas decreases as E/N and the average electron energy increase. This behavior is consistent with the $1/\epsilon$ energy scaling of the Coulomb-like effective cross section of the Debye-shielded dust particles. The effect of dust parameters on transport coefficients is shown in Fig. 3. Ionization rate coefficients of Ar as a function of dust density are shown in Fig. 3(a) for two E/N values. These coefficients have been normalized by their pristine values to emphasize the effect of the dust. Increased dust densities result in lower overall rates of ionization as a result of the progressive reduction of the high-energy component of the EED. The threshold dust density for which this effect occurs, 10^3–10^5 cm^{-3}, decreases with decreasing

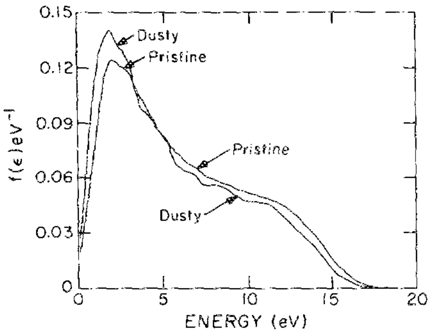

FIG. 1. Electron energy distribution for electron swarms in pristine argon and argon contaminated with particles 1 μm in radius having a density of $\rho = 10^5$ cm^{-3}. The reduction in high-energy electrons in the dusty plasma is due to scattering from the Debye shield surrounding the particles. The discharge conditions are $E/N = 7$ Td (1 Td $= 10^{-17}$ V cm^2), $P = 0.1$ Torr, $T_{\rm gas} = 300$ K, and $n_e = 5 \times 10^{11}$ cm^{-3}.

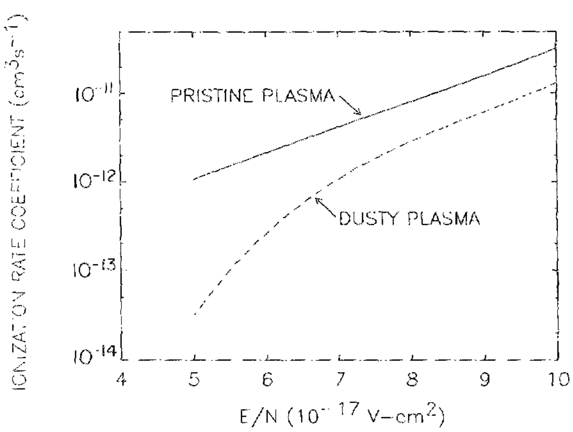

FIG. 2. Rate coefficient for ionization of Ar as function of E/N for the same discharge conditions as in Fig. 1. The decrease in the rate coefficient in the dusty plasma is due to the reduction in the density of high-energy electrons in the EED.

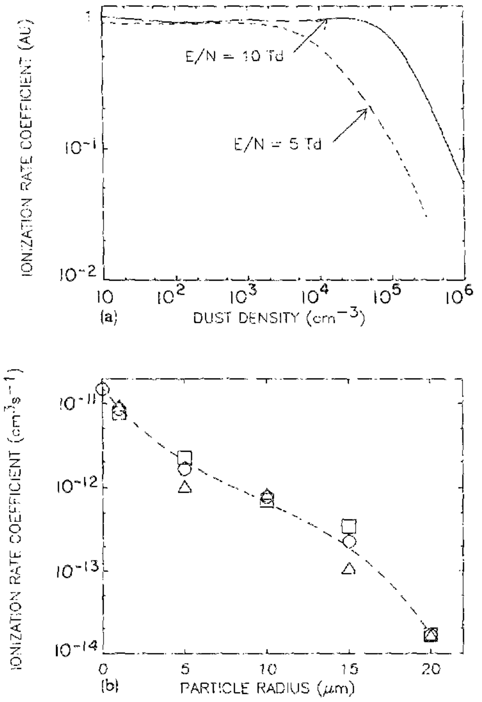

FIG. 3. (a) Rate coefficient for ionization of Ar as a function of dust density for $E/N = 5$ and 10 Td (1 Td $= 10^{-17}$ V cm^2). These coefficients have been normalized by their pristine values. Note that the difference between the pristine and dusty plasmas is more severe at lower E/N. (b) Rate coefficient for ionization of Ar as a function of the radius of the dust particles.

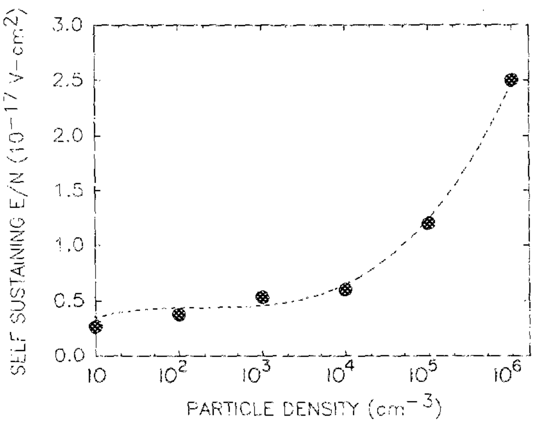

FIG. 4. Self-sustaining electric field/number density $(E/N)_s$ as a function of dust density ρ. $(E/N)_s$ increases as ρ increases due to a reduction in the density high-energy electrons, which reduces the rate coefficient for ionization. Discharge conditions are $P = 0.1$ Torr, $T_g = 300$ K, and $n_e = 5.0 \times 10^{11}$ cm^{-3}. The diffusion length is based on a wall separation of 4 cm.

E/N. The effect of the size of the dust particle on k_I is shown in Fig. 3(b). The decrease in k_I at larger dust radii is due to both the scattering from the Debye shield around the particle and from physical obscuration.

In order to evaluate the effect of particulate contamination on the operation of a glow discharge, we calculated the change in the self-sustaining electric field/number density, $(E/N)_s$, which results from their presence. To gauge this effect, $(E/N)_s$ was calculated using a simple model for a steady-state positive column,

$$\frac{1}{n_e}\frac{\partial n_e}{\partial t} = k_I(E/N,\rho)N - \frac{D_a}{\Lambda^2}(E/N,\rho) = 0, \qquad (4)$$

where N is the neutral gas density, D_a is the ambipolar diffusion coefficient, ρ is the dust density, Λ is the electron diffusion length, and k_I is the ionization rate coefficient.[15] The effect of dust density on $(E/N)_s$ is shown in Fig. 4. Increasing dust density causes a shift in $(E/N)_s$ to larger values to account for the decrease in k_I. This effect is also a function of other parameters such as discharge dimensions and gas pressure.

In conclusion, a hybrid Monte Carlo/molecular dynamics computer simulation of low-temperature plasmas has been used to investigate the effect of contamination of otherwise pristine argon plasmas with particulates of $\geqslant 1$ μm radius. The presence of particulates causes a reduction in the number density of high-energy electrons and reduces the electron impact rate coefficients for processes with high threshold energies, such as ionization. The self-sustaining E/N for plasmas contaminated with particulates increases for this reason. The effect of particulate contamination on electron transport coefficients is proportional to particulate density, with a threshold of about 10^3–10^5 cm^{-3} for gas pressures $\leqslant 1$–10 Torr.

This work was supported by the National Science Foundation (CBT 88-03170) under the direction of Dr. R. Goulard and Dr. W. Grosshandler.

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