Monte Carlo-fluid hybrid model of the accumulation of dust particles at sheath edges in radio-frequency discharges

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Particulate contamination (dust) has been observed to accumulate near the sheath-plasma boundary in both radio-frequency (rf) and direct-current (dc) discharges. We have developed and applied a hybrid Monte Carlo-fluid simulation of electron, ion, and charged dust transport in rf discharges to investigate the dynamics of particulate contamination. The processes governing the transport of charged dust in the model are drift of partially shielded particles in the electric field, collisions with the fill gases, and viscous ion drag arising from Coulomb interactions of particles with ions drifting and diffusing in the plasma. We find that negatively charged dust particles accumulate near the sheath-plasma boundary, and that transport of the particles is dominated by ion drag.

Particulate contamination ("dust") has been experimentally observed in both direct-current (dc) and radio-frequency (rf) glow discharges.[1-9] It is known that such contamination can alter electron transport in low pressure plasmas[10] and lead to instabilities in the flow of the plasma.[11] Dust is problematic in plasmas used for fabrication of microelectronic components because it can contaminate the wafer and reduce production yields.[12] Experimental observations of dust in low pressure parallel-plate rf discharges using laser light scattering have shown that particles preferentially accumulate near the sheath-plasma boundary and can form complex structures.[1-9] In this letter we present a model of charged particle dynamics (electrons, ions, and charged dust) in parallel-plate rf discharges using a hybrid Monte Carlo-fluid simulation. We find that dust particles (modeled as massively large multiply charged negative ions) accumulate near the sheath edges, and that particle transport is dominated by Coulomb interactions with ions that are diffusing and drifting in the plasma ("viscous ion drag").[13]

Our hybrid model for electron, ion, and charged particle transport combines a Monte Carlo simulation (MCS) and a fluid simulation (FS). The MCS is used to generate nonequilibrium source functions for electron impact processes and electron transport coefficients, and is functionally equivalent to that described in Refs. 14 and 15. These source functions and coefficients are then used in the FS from which we obtain the species densities and electric field. In the FS, the continuity equations for all charged species are integrated, and Poisson's equation is solved for the electric field. The density of species n is obtained in one spatial dimension z from the continuity equation:

$$\frac{\partial n}{\partial t}= - \frac{\partial}{\partial z}\left(v_d n - D\cdot\frac{\partial n}{\partial z}\right) + S, \qquad (1)$$

where S is the source for ionization or attachment, v_d is the drift velocity and D is the diffusion coefficient, all obtained from the MCS. The source terms reflect the full nonequilibrium nature of the electron energy distribution. Tabulated transport coefficients as a function of the reduced electric field E/N are used for the ions.[16] Mobilities and diffusion coefficients are applied within the FS using the local field approximation. The FS is structured so that an arbitrary number of positively and negatively charged species having different transport coefficients can be simulated. The continuity equations are couched in finite difference form using the donor cell method, and both the continuity equations and Poisson' s equation (using electron, ion, and dust charge densities) are directly integrated using the Euler method. Ion charge exchange reactions are included in the source terms, as well as ion-ion recombination and electron-ion recombination on particulate surfaces.

The method of solution in the hybrid model is to run the MCS for 10–20 rf cycles to generate the source terms and transport coefficients. The FS is then run for 10–20 rf cycles to obtain the electric field $E(z,t)$ which is used in the next run of the MCS. Convergence, based on total plasma density, is obtained after 100's to 1000 rf cycles. A single case typically requires 2–10 h on a laboratory minicomputer (Stardent 3000).

Dust particles are simulated as massively large multiply charged negative ions having a discharge averaged density of 10^3 cm^{-3} and diameters of 10's nm to 10 μm. Neutral particles of this size quickly become negatively charged and a space charge sheath forms around them. The actual number of negative elementary charges Z_D on dust particles is uncertain. Experimental estimates range from[17] Z_D= 10–100's to the field emission limit,[18] $Z_D \simeq 10^5$.

We have derived a simple expression for Z_D for use in the model by equating the flux of electrons to a particle with the flux of positive ions. The flux of electrons at the surface of the dust is related to the electron flux outside the particle's sheath through the Boltzmann relation.[19] The classical Coulomb scattering trajectory[20] is then used to find the impact parameters for which electrons and ions hit the particle. Assuming that the sticking probability is unity and that electrons and ions approach at their thermal speed, one obtains

$$Z_D = \frac{6\pi\epsilon_0 r_D (1-p) k_B T_e T_+}{e^2 (pT_e + T_+)}, \qquad (2)$$

where $p = n_+/n_e (T_+/m_+)^{1/2} (T_e/m_e)^{-1/2}$, k_B is Boltzmann's constant, e is the unit charge ($e > 0$), and ϵ_0 is the permittivity of free space (MKS). n, m, and T are the density, mass, and temperature, and the subscripts e, $+$, $-$, i, and D refer to electrons, positive ions, negative ions, ions of either charge, and dust, respectively. Both n and T are functions of z. Equation (2) implies that there are 100's –1000 elementary charges on a 1-μm-diam particle under typical processing plasma conditions. It is the unshielded charge Z_D which appears in Poisson's equation. It is likely that steady-state analyses of the particle charge [such as Eq. (2)] underestimate Z_D in regions where densities and temperatures rapidly vary, such as the sheaths.

We consider three forces on the charged dust in the model. First, we assume that the Debye shielding around the particle is not perfect and that the particle experiences an electrostatic force as though it were an ion of mass m_D with Z'_D negative elementary charges. Second, the particle is slowed by momentum transfer collisions with the fill gases. The third is a viscous drag arising out of the directed motion of the ions. A polarization force may be important for segregating very large particles (>10 μm) in the sheaths, and thermophoretic forces may influence dust transport,[21] but these effects are not considered here.

The shielded charge Z'_D varies according to the local electric field and plasma conditions. In weak field regions such as the bulk plasma, Debye shielding is quite effective, and we assume $Z'_D = 1$. Debye shielding is less effective for particles in the strong rf fields in the sheath because the response times for electrons and ions in the Debye sphere (μs–ms) are much longer than the rf period (≈ 75 ns). Particles in the sheaths may therefore respond to the electric field as though they are less than fully unshielded ($1 \leqslant Z'_D \leqslant Z_D$). To account for this effect we compare the macroscopic electric field $E(z,t)$ to the microscopic field E_m at a distance of one Debye length λ from a dust particle of charge Z_D, $f = |E(z,t)/E_m|$. (f is limited so that $0 < f \leqslant 1$.) The charge which is used to obtain the drift of the particle in the macroscopic field is then

$$Z'_D = (1-f)1 + fZ_D. \qquad (3)$$

The viscous ion drag forces arise from momentum transfer Coulomb collisions with ions having directed fluxes.[22] A particle will experience (on average) an impulse $m_i v_i$ for each ion having velocity v_i which passes within roughly a Debye length. The viscous drag cross section σ_{iD} can be approximated by using the radius for 90° Coulomb collisions[13,23] for ions scattering from a particle with charge Z_D. To allow a smooth transition between the bulk plasma (where the particle is shielded on a scale length determined by electrons) to the sheath (where the scale length for shielding is determined by positive ions) we approximated that $\lambda = (2n_e\lambda_e + n_+\lambda_+)/(n_e + n_+)$, where $\lambda_s = [\epsilon_0 k_B T_s/(n_s e^2)]^{1/2}$ for species s. In plasmas having many ion species σ_{iD} is a density weighted average value. The drift velocity for charged dust used in the continuity equation is then obtained by balancing the electrostatic and viscous drag forces,

$$v_D = \mu_D \cdot \left(-Z'_D \cdot E + \sum_i n_i \sigma_{iD} m_i v_i |v_i| \right), \qquad (4)$$

$$v_i = \mu_i E - D_{iN} \frac{1}{n_i} \frac{\partial n_i}{\partial z}, \qquad (5)$$

where the sum is over all ions, μ_i is the ion mobility, and D_{iN} is the ion-neutral diffusion coefficient.

We have modeled dust dynamics in symmetric rf (13.56 MHz) parallel-plate discharges for a variety of fill gases and conditions. Since we do not presently address the dynamics of particle production, cases are started with a spatially uniform distribution of dust and the density is

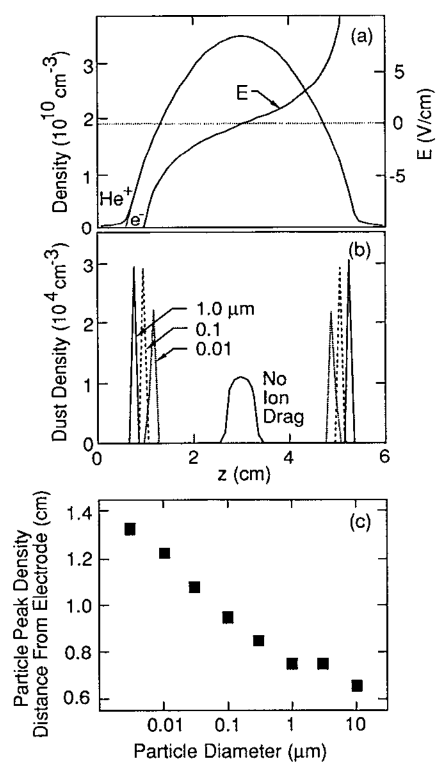

FIG. 1. Results from the hybrid model for a 200 mTorr He rf discharge. The applied voltage amplitude is 500 V (13.56 MHz). (a) Pristine He discharge. Instantaneous electron and ion densities and electric field for the phase of the cycle where the left ($z = 0$) electrode is most cathodic. (b) Continuation of the same calculation after adding particles of various sizes initially having a uniform density $n_D = 1 \times 10^3$ cm^{-3}. The density of 0.5 μm particles is also shown when the ion drag force is removed from the calculation. (c) The distance of the peak dust density from the nearest electrode as a function of particle diameter.

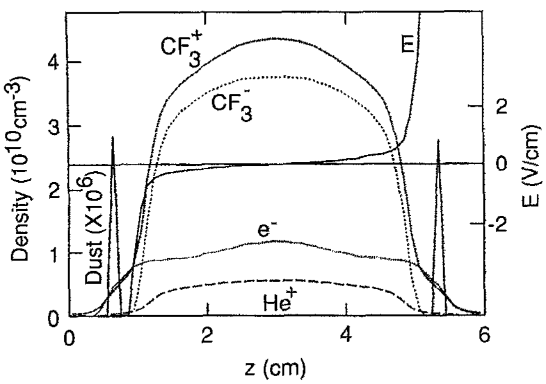

FIG. 2. Results from the hybrid model for a contaminated 200 mTorr $He/CF_4 = 95/5$ rf discharge. The applied voltage amplitude is 200 V (13. 56 MHz), and the particle diameter is 1 μm. (a) Electron and ion densities, and electric field; (b) particle density. To minimize the number of species we assumed all attachment results in CF_3^-.

allowed to evolve. Results from the model for dust having several diameters in a 200 mTorr He discharge are shown in Fig. 1. The dust density maxima occur where the ion drag force, which is directed toward the electrode, is balanced by the confining electrostatic force provided by the shielded negative particle charge Z_D'. The location at which the forces balance depends upon the particle size because the magnitude of the ion drag force depends upon the particle charge Z_D, which in turn depends upon the particle radius r_D [Eq. (2)]. The model predicts that particles segregate by size, with larger particles residing closer to the electrode, in agreement with experimental observations.[3,21]

Dust does not generally accumulate at the midplane of the discharge where the time-averaged plasma potential is most positive. Particles are swept away from the midplane because the net ion drag force due to ion transport to the electrodes is greater than the confining force $-Z_D'E$. We can test our assumptions for the shielded particle charge Z_D' and the ion drag force by varying these effects in the model. When Z_D' is not allowed to vary [$f=0$ in Eq. (3)], the confining electrostatic force is reduced and dust accumulates within the sheath. If ion drag is removed entirely from the model, all particles collect at the midplane of the discharge [as shown in Fig. 1(b)] where the time-averaged plasma potential is most positive. Both of these outcomes are contrary to experimental observations.[1–9,21]

Results for a discharge in a 200 mTorr $He/CF_4 = 95/5$ gas mixture are shown in Fig. 2. Because the ionization threshold for He is larger than that for CF_4, the ions consists of nearly equal densities of CF_4^+ and CF_3^- ions, with a much smaller density of He^+. The character of the ion drag force is therefore distinctly different than in a pure He discharge where there is only a single ion species. Particle behavior in this gas mixture, though, is very similar to that observed in pure He, with the particle density peaking near the sheaths at the location where the ion drag and the confining electrostatic forces balance. Although experiments suggest that a trapping potential exists at the same location where the particle density peaks,[2] and such potential wells almost certainly exist in some electronegative discharges, our present results indicate that the wells are not a prerequisite for particle accumulation.

In conclusion, we have developed a hybrid Monte Carlo-fluid simulation for charged particle transport in rf discharges. Transport of the particles is dominated by Coulomb interactions with ions diffusing and drifting out of the plasma (ion drag) and by electrostatic forces in regions where Debye shielding is not effective. Dust tends to accumulate where the ion drag and the electrostatic force due to the unshielded charge balance.

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