Numerical Simulations of Nanodusty RF Plasmas
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Abstract—Results are presented of numerical simulations of the spatiotemporal evolution of nanoparticles that nucleate and grow in a nonthermal radio-frequency plasma. This paper highlights the effects of several operating parameters.

Index Terms—Dusty plasma, numerical simulation.

Nanoparticles that nucleate and grow in low-temperature plasmas are of interest both as an unwanted source of contamination in semiconductor processing and as a deliberate synthesis product for applications such as photovoltaics, catalysis, and cancer treatment. In either case, the appearance, growth, and charging of nanoparticles can profoundly affect the plasma, leading to a rich spatiotemporal evolution with many features that are not well understood.

Results are presented of numerical simulations of a parallel-plate capacitively coupled radio-frequency (RF) plasma in which several operating parameters are varied, including pressure, gas temperature, temperature gradient, RF voltage, and gas velocity. These parameters are varied one at a time from base case conditions of 13.33 Pa (100-mtorr) pressure, 300 K gas temperature with zero temperature gradient, 100-V RF voltage amplitude, zero gas velocity, 13.56-MHz frequency, and a 3-cm electrode gap, with the upper (showerhead) electrode being powered and the lower one grounded. The plasma is assumed to consist of pure argon, while the nanoparticles are assumed to have the properties of silicon.

These simulations use a modified version of a code developed by Girshick et al. [1], [2], which couples a 1-D fluid plasma model to a solution of the aerosol general dynamic equation that employs a sectional representation of the particle size distribution function. Population balance equations are solved for electrons and ions, together with the electron energy equation and Poisson’s equation for the electric field. As chemistry is not modeled, nucleation and surface growth rates are treated as free parameters of the simulation. Particle charging is modeled using orbital motion limited theory. Size- and charge-dependent particle coagulation is calculated, accounting for the effect of image potentials in coagulation between charged and neutral particles. Nanoparticle transport effects considered include motion due to electrostatic forces, Brownian diffusion, neutral drag, ion drag, gravity, and thermophoresis.

Fig. 1 shows simulation results for the spatial distribution over the electrode gap of the particle size distribution function $dN/d\log(d_p)$, with $N$ being the particle concentration and $d_p$ being the particle diameter. White lines represent isocontours of the mean value of the net negative charge per nanoparticle. Each row shows results for a given set of conditions at three different times (0.5, 1.5, and 3.0 s) following the onset of particle nucleation. Each column shows results at a given time for each of the six different sets of conditions.

The first row of figures shows results for the base case. At 0.5 s, most particles are in the 5–10-nm diameter range. Negatively charged nanoparticles are pushed to the center of the plasma by the electric field, while neutral and positively charged particles diffuse outward and are lost before they have time to grow beyond a few nanometers in diameter. At 1.5 s, a pronounced “V” has developed in the spatial distribution of nanoparticles, caused by ion drag, which pushes particles out of the center and toward the electrodes. At 3 s, the outward spread of the particle cloud has quenched nucleation. Any remaining nonnegative particles are then either lost by diffusion or scavenged by the larger particles. The spatial asymmetry seen in the nanoparticle distribution at 3 s is caused by gravity, which, for particle diameters around 50 nm, is a small but not negligible effect.

When the pressure is doubled (second row), fewer nanoparticles are lost by Brownian diffusion before being charged and pushed toward the center. When the gas temperature is increased from 300 K to 500 K (third row), the outward spreading of the particle cloud is reduced. This is primarily due to a reduction in ion drag, caused by an increase in the thermal component of ion motion. The introduction of a temperature gradient (fourth row) drives thermophoresis, which pushes nanoparticles toward the colder (upper) electrode. When the RF voltage is doubled (fifth row), the ion drag force increases, causing nucleation to be quenched sooner by the spreading particle cloud. The introduction of gas flow through the upper (showerhead) electrode (sixth row) pushes particles toward the lower electrode. A surprising consequence is that the nanoparticle concentration at 1.5 s is greater close to the showerhead than near the lower electrode. This effect is caused by the quenching of nucleation by the downward-spreading nanoparticle cloud.

REFERENCES

Manuscript received November 30, 2010; accepted May 7, 2011. Date of publication June 16, 2011; date of current version November 9, 2011.

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Digital Object Identifier 10.1109/TPS.2011.2157945
Fig. 1. Results of numerical simulation for spatial distribution of particle size distribution function for different operating conditions. Top row shows base case, while each subsequent row varies one operating parameter at a time, as indicated. White contour lines represent average negative charge per nanoparticle.