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Microscopic Particle Simulation of Air Ionization

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Abstract

A two-dimensional electrodynamic model is developed from first principles to study the microscopic physics of gas breakdown and charge transport in corona devices. The model uses a combination of particle simulation for atomic processes and macroscopic rate equations for the various chemical reactions. The particle model alternates between field solve and particle push. A Monte Carlo collision model is used to describe energy-dependent electron-neutral collisions including elastic collisions as well as ionization and dissociation processes. A novel "particle clumping" technique is introduced in phase space to create super-particles, thus restricting the total number of simulation particles to within a tractable number of a few million. It is shown that the model may be useful for studying microscopic air ionization physics, including the production of ozone. Some preliminary results are presented for both positive and negative DC corona devices.

Introduction

Corona devices are useful for many applications including copiers and electrostatic printers, plasma etching systems, plasma display panels, electrostatic precipitators, electronic cooling devices, and surface pre-coating agents. Except for plasma etching and plasma displays, all the other applications involve low temperature and high-density plasmas in weakly ionized gases at atmospheric pressure. Much of the understanding of the physical chemistry of corona discharge in air has been derived from empirical studies. There has been little prior work in the quantitative simulation of this phenomenon from first principles. Traditional plasma simulations address partial vacuum or very low-pressure conditions. At atmospheric pressure, collisions with neutrals are much more frequent and lead to exponential increase over time of the total particle count, resulting in a significant increase in overall problem size. In addition, the number of charge species is also much more diverse. Air ionization incurs a multitude of charged species, all contributing to collisional interactions and chemical reactions. Any simulation model must necessarily include the primary and secondary species and other less desirable by-products such as ozone, which contributes to environmental pollution.

In this work, we present a two-dimensional numerical model derived from first principles to simulate the microscopic physics of corona discharge. The model uses a combination of Particle-in-Cell (PIC) simulation for atomic processes and macroscopic Chemical Rate Equations (CRE) for the various species reactions. The particle model alternates between field solve and particle push. A Monte Carlo Collision (MCC) model is used to describe energy-dependent electron-neutral collisions including elastic collisions as well as ionization and dissociation processes. A novel "particle clumping" technique is introduced in phase space to create super-particles, thus restricting the total number of simulation particles to within a tractable number of a few million. It is shown that the model may be useful for studying microscopic air ionization physics, including the production of ozone. Some preliminary results are presented for both positive and negative DC corona devices.

Simulation Algorithm

The particle simulation method is a natural choice to study the microscopic physics processes in the corona device as it lends itself intuitively to the description of electron dynamics in the corona discharge. Newton's equation of motion is followed in time as it evolves in velocity and position space (phase space) to determine the trajectory of each particle. The PIC model is used to calculate the force on a particle and the charge on a grid point.[1] The equation of motion for a particle is given by:

$$m\, d\mathbf{v}/dt = q\mathbf{E} \qquad (1)$$

where m and q are the mass and the charge of the particle, **v** is its velocity and **E** is the electric field at the particle location. The electrostatic Coulomb force on a particle is calculated from the electric field by solving Poisson's equation. The particle simulation method alternates between a field solve and a particle push at each time-step. The field solve can be accomplished by any of several traditional approaches including analytic and numerical schemes.[2] A particle-mesh[3] approach is implemented to reduce overall computation size where the fields are computed on a mesh and interpolated to particle centroids for tracking. In N

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particle systems, the order of computation is NM, for a mesh of size M, compared to N^2 for particle-particle interactions where N >> M.

Monte Carlo Collisions

The Monte Carlo Collision model[4,5,6] can describe particle-particle events in which random numbers are used to determine collision outcomes. In this method, the probability of a collision of the i-th particle with the background neutral gas in a time step Δt is given by:

$$P_i = 1 - exp\ [\ -n_0\ \sigma(E_i)\ v_i\ \Delta t\ ] \quad (2)$$

where n_0 is the neutral gas number density, $\sigma(E_i)$ is the total collision cross-section which in general depends on the kinetic energy of the i-th particle and v_i is the velocity of the particle relative to the velocity of the neutral gas. Therefore, $n_o\sigma$ is a measure of the number of collisions per unit length, $1/n_o\sigma$ is the mean free path between collisions, and $n_o\sigma v_i$ is the collision frequency. The cross-section in (2) is the total cross-section, representing multiple ionization and dissociation collisions. From (2) it is clear that for $\Delta t = 0$, $P_i = 0$ and for $\Delta t = \infty$, $P_i = 1$ meaning the particle will eventually collide with neutrals. For a finite Δt, the probability of a collision is determined by comparing P_i with R_i, representing a uniform random number between 0 and 1. For $P_i > R_i$, the particle i is assumed to have sustained a collision within the time step Δt. The emergent velocities of the particles associated with the collision are computed from energy considerations. For an electron-neutral ionization collision, the energies of the two electrons after the collision must be equal to the energy before the collision minus the ionization energy. The incident energy is divided into the scattered and the created electrons whose magnitude as well as the scattering angles are calculated from a model differential cross-section associated with the collision.[6] Dominant electron-neutral collision processes are given by

$$O_2 + e = O_2^+ + e + e \quad (3)$$

$$N_2 + e = N_2^+ + e + e \quad (4)$$

$$H_2O + e = H_2O^+ + e + e \quad (5)$$

$$O_2 + e = O + O + e \quad (6)$$

where H_2O in (5) represents the effect of the water humidity in air which may have a significant effect on the corona discharge.[7] The dissociation of O_2 molecules in (6) leads to generation of ozone molecules. Collision cross-sections for the reactions, (3)-(6), are published in the literature.[8] At atmospheric pressure, the neutral density is very high at $2 \times 10^{19}/cm^3$ so that the electrons and neutral gas suffer frequent collisions. Electrons can gain energy of more than a few electron volts (eV) because of the very large potential gradient near the wire leading to formation of the high-energy electron tail.

Macroscopic Chemical Rate Equations

In addition to the atomic processes between the electrons and neutral gas described by (3)-(6), there are many other ion and molecular reactions taking place in the corona device.[9] Reactions included in the present simulations are

$$O + O_2 + O_2 = O_3 + O_2 \quad (7)$$

$$O_2 + O_3 = 2O_2 \quad (8)$$

$$H_2O^+ + H_2O = H_3O^+ + OH \quad (9)$$

$$O_2 + O_2 + e = O_2^- + O_2 \quad (10)$$

$$O_2^+ + O_2^- = 2O_2 \quad (11)$$

where (7) and (8) describes the generation and recombination of ozone molecules, (9) represents hydrate generation, and (10) and (11) denotes the generation of negative O_2^- ions and their recombination.[10] The processes are followed using these chemical rate equations and coefficients with densities of atomic O, H_2O^+, electrons, O_2^+ and N_2^+ calculated from the particle model.

Particle Clumping

Because of rapid ionization, the number of electrons and ions in each run increases exponentially in time so that the problem becomes intractable due to sheer size. A novel method called, "particle clumping", is used to combine two or more particles that occupy the same phase space into a super-particle. This scheme limits the overall size of the simulation from exceeding a few million particles. Conservation of charge and mass are trivially satisfied by enforcing the charge and mass of the super-particle to be the algebraic sum of the original particles. Conservation of energy and momentum cannot be met precisely at the same time except for a trivial case when the two particles are coincident in phase space. Since the plasma is highly collisional with neutral gas, the electron energy and the momenta are not conserved, as they are constantly lost to heat the neutral gas. This fact allows particle clumping to continue so long as the numerical error due to clumping remains much less than the intrinsic loss of energy and momentum due to the interaction of the electrons and the neutral gas. More explicitly, numerical diffusion due to clumping must remain much smaller than the diffusion caused by the collisions of the electrons with the neutral gas. Mathematically this condition[5] is $(\delta v)^2/\tau_{clump} << D_{collision}$ where δv is the size of the velocity space cell and τ_{clump} is the time step between clumping actions.

Assumptions and Initial Conditions

A typical configuration for a corona device is the scorotron, shown in Fig. 1, where a high voltage wire creates a discharge between the biased three-sided shield and the bottom grid over the charging substrate. The shield is typically grounded and the grid is biased at a potential of

the same polarity as the wire to select the desired level of charging. A 128 x 128 two-dimensional computational grid is overlaid on top of the cross-section within the area included by the shield and grid. The high voltage wire is located at the center of the included area and may be biased at +5 kV (positive wire) or -5kV (negative wire) potential. The corresponding grid potential is +1 kV or -1 kV to control the ion flux to the photoreceptor. The grid is modeled as alternating conductor-air spaces at 50% duty cycle with a spatial pitch of approximately 1 mm. The model is normalized by the grid size in length and by the electron plasma frequency, $1/\omega_{pe}$, in time. Therefore, for the assumed initial electron density of $10^{10}/cm^3$ and temperature of 1 eV, the Debye length is equal to 7.43×10^{-3} cm, or a linear dimension of 0.95 cm. To represent the initial electrons, oxygen and nitrogen ions, 512 x 512 simulation particles are used for each species distributed across the 128 x 128 spatial grid. Since the initial electron density is assumed to be $10^{10}/cm^3$, each simulation electron represents 1.7×10^4 real electrons. The time step of integration is chosen to be $0.0025/\omega_{pe}$, which corresponds to 0.44 picoseconds. This small time step is needed since the collision mean free path is very small due to the high neutral air density. The typical ionization cross-section is a fraction of 10^{-16} cm^2 for the reactions described by (3)-(6). The neutral gas density n_o is $2 \times 10^{19}/cm^3$ and the electron velocity v is about 10^8 cm/s so that the collisional ionization frequency is given by $n_o\sigma v=10^{11}/s$, suggesting that the initial plasma density can double in 20 picoseconds. The mean free path for the ionization is given by $1/n_o\sigma=0.5 \times 10^{-3}$ cm.

Results and Discussion

A two-phase procedure is used for each simulation run. The first phase proceeds through 3000 time-steps corresponding to 1.32 nanosecond in real time at which point the total number of particles exceeded 6 million. Then particle clumping is invoked to reduce the particle count to no more than 2 million. The second phase continues execution through 10000 time-steps, corresponding to 4.4 nanoseconds in real-time. The clumping is found to be numerically very stable. Comparison of velocity distributions before and after clumping indicates that the two sets are quite similar, assuring the continuity of the same physical processes. The initial electrostatic potentials are determined by solving Laplace's equation since the space charge is electrically neutral. The surface potential plot for the positive wire case is shown in Fig. 2. The potential peak indicates the high electric field in the vicinity of the wire. Fig. 3 shows the scatter plot for primary electrons for the negative wire case. The distribution is fairly homogeneous except for the low-density hole near the wire caused by the outward repulsion of electrons. Fig. 4 shows the secondary electron scatter plot for the negative wire case. The distribution is localized around the wire and the two lower shield-edge corners where high fields sustain local ionization. The distribution near the wire shows twin peaks straddling the wire with a density of $2.4 \times 10^{13}/cm^3$. Corresponding scatter plots for atomic O, H_2O^+, N_2^+, and O_2^+, exhibit qualitatively similar distributions. Ozone generation in (7) uses the rate coefficient of 6.5×10^{-34} $cm^6/molecules^2 second$.[13] The peak density of ozone for the positive wire case is about $1.2 \times 10^8/cm^3$ and their spatial distribution is again similar to that of the secondary electrons with twin peaks around the high voltage wire, as shown in Fig. 5. The time history for ozone density generation near the center of the corona device for positive (solid) and negative (dashed) wires are shown in Fig. 6. At the end of the run (4.4 nanoseconds), the density for the positive wire case is 10 times larger than for the negative wire. However, this is due to the distribution being more concentrated near the center for the positive wire. In fact ozone density contours at 4.4 nanosecond show that both densities are about the same at about $2.5 \times 10^{11}/cm^3$. The ozone density initially increases rapidly but slows at around 4000 steps and increases much more slowly thereafter.

Concluding Remarks

A two-dimensional hybrid PIC-MCC-CRE model was derived from first principles to simulate air ionization. A novel "particle clumping" technique was introduced in phase space to create super-particles, thus restricting the total number of simulation particles to within a tractable number of a few million. It was shown that the model might be useful for studying microscopic air ionization physics, including the production of ozone. Some preliminary results were presented for both positive and negative DC corona devices at nanosecond time scales. Some future refinements include improve ion chemistry, optimized computational grid, and application to a specific device to validate the model for design extrapolation.

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Biography

Meng H. Lean is a Principal Scientist with Xerox Corporation, Wilson Center for Research & Technology. Dr. Lean is involved in simulation from first principles of marking processes and systems. His work emphasizes development of numerical tools, incorporating innovative hybrid numerical algorithms, to guide and optimize the design and fabrication of printing systems hardware.

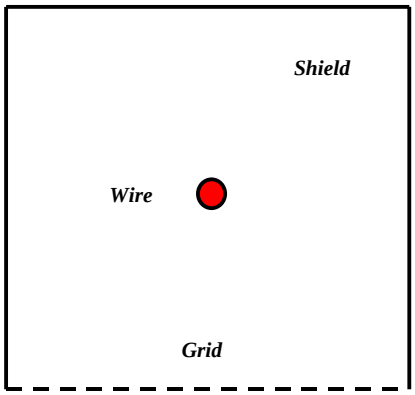

Figure 1. Schematic cross-section of the scorotron device.

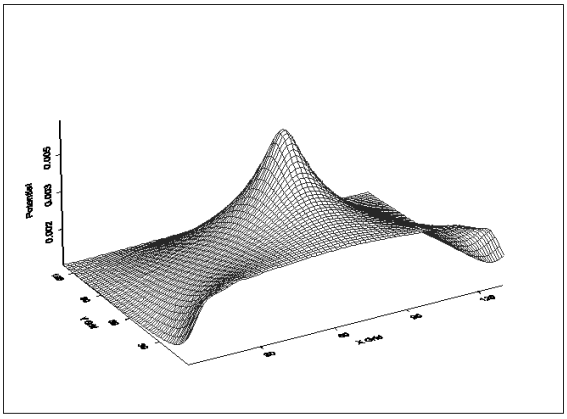

Figure 2. Surface potential plot for positive wire.

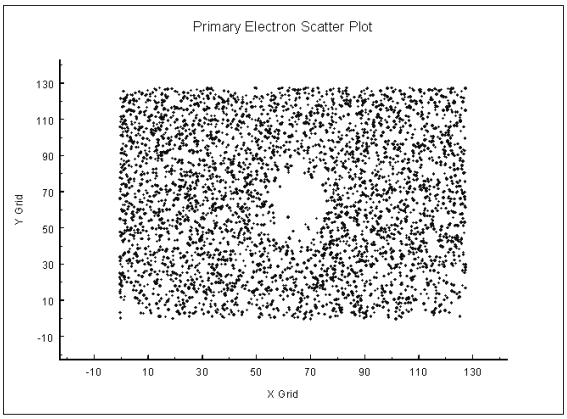

Figure 3. Primary electron scatter plot for negative wire.

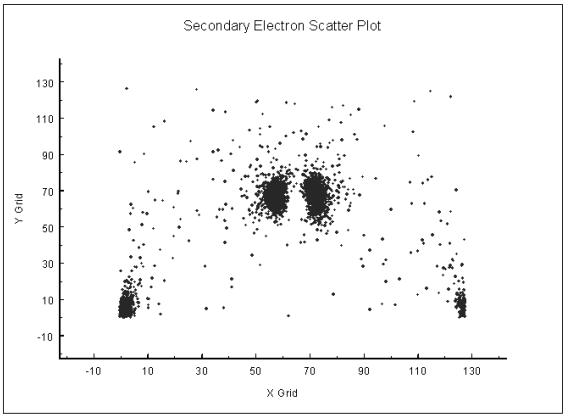

Figure 4. Secondary electron scatter plot for negative wire.

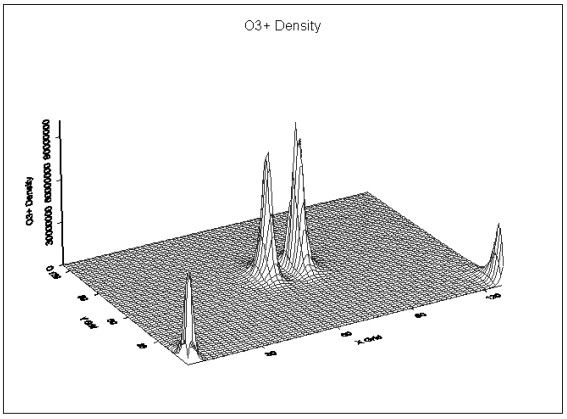

Figure 5. Surface plot of ozone density for positive wire.

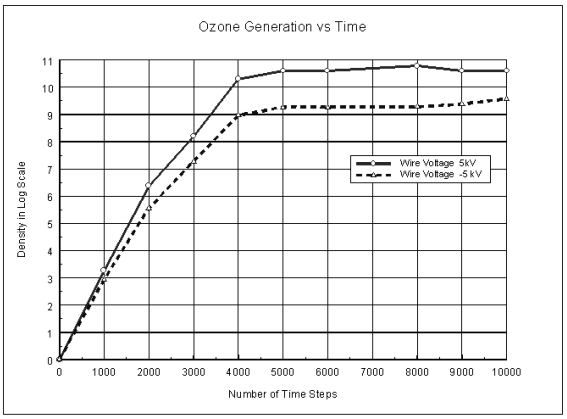

Figure 6. Time history for ozone generation.