A 2-D Self-Consistent DSMC Model for Chemically Reacting Low Pressure Plasma Reactors

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Abstract

This paper will focus on the methodology of using a 2D plasma Direct Simulation Monte Carlo technique to simulate the species transport in an inductively coupled, low pressure, chemically reacting plasma system. The pressure in these systems is typically less than 20 mtorr with plasma densities of approximately 10^{17} #/m^3 and an ionization level of only 0.1%. This low ionization level tightly couples the neutral, ion, and electron chemistries and interactions in a system where the flow is subsonic. We present our strategy and compare simulation results to experimental data for Cl$_2$ in a Gaseous Electronics Conference (GEC) reference cell modified with an inductive coil.

1 Introduction

The microelectronics industry is pursuing low pressure etch and deposition reactor systems to achieve the uniformity which is required to meet the demands of gigascale integrated circuits. These reactors operate at pressures < 50 mtorr and the high density plasma etch reactors have plasma densities ~ 10^{17} - 10^{18} #/m^3 in an effort to achieve a high etch rate and anisotropic etching. Low gas pressures are necessary to ensure collisionless ion transport through plasma sheaths, which provides anisotropy in the ion flux directed toward the surface. This enables etching of high aspect-ratio features.

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The collisional mean-free-path at 1 mtorr is 100 mm and the diameter of the wafer is 200 mm; therefore, a continuum treatment of the neutral and ion transport is suspect. The Direct Simulation Monte Carlo (DSMC) technique [1], a kinetic simulation of the Boltzmann equation, is an ideal method for these processes since it has the capability of correctly modelling the high density inlet, the complex gas phase chemistry, and the multi-species transport in the system.

These reactor systems are composed of three distinct time constants for the transport of neutrals, ions, and electrons and as such have not been previously modelled in a kinetic, self-consistent manner for these low pressures. The focus of this paper will be on presenting a self-consistent kinetic code for a low pressure chemically reacting plasma and comparing predictions with experimental data. We use a 2D DSMC code, Icarus, which has been optimized for the massively parallel computing environment [2] to transport the neutrals and ions. The Variable Hard Sphere model is used to determine neutral-neutral and neutral-ion collision frequencies. A Coulomb interaction model is used for ion-ion collisions. Gas phase chemistry can be modeled with either a kinetic approach (steric factors) or a continuum-like treatment (reaction rates). Surface chemistry is modelled with either a simple reaction probability (sticking factor) or a site coverage dependent model. There are two major plasma modelling strategies for these systems: 1) model kinetic electrons, ions, and neutrals with a Poisson field solver, and 2) model kinetic ions and neutrals and assume quasi-neutrality with an ambipolar field model. The first is a direct method with little assumptions; however, the multiple time constants of this system make this strategy computationally intractable for all but the simplest systems. We present the second approach in this paper.

2 Quasi-Neutral Plasma DSMC Model

The characteristics of our plasma DSMC model are:

a) DSMC kinetic treatment for neutrals and ions,

b) local quasi-neutral assumption for cell average electron density,

c) ambi-polar fields: $E = \nabla(kT_e n_e) / (q n_e)$

where T_e, n_e, and q are electron temperature, density and charge,

d) 1D sheath model of Riley[3] to determine positive ion surface energy,

e) Inductively Coupled Power (ICP) determined from ORMAX [4],

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f) control volume electron energy equation:

$$\frac{\partial}{\partial t}((3/2)n_e k T_e) + \nabla \cdot q_e + 3\frac{m_e}{m_n}n_e k \nu_m (T_e - T_g) + P + \sum_j H_j R_j = 0$$

where q_e is the electron heat conduction $= -\lambda_e \nabla T_e$

with $\lambda_e = 2.4\ k^2\ n_e\ T_e\ /\ (m_e\ \nu_{eH})$ [ref 5 - for partially ionized gas],

P is the ICP power (item e), and

$H_j R_j$ are the heat of formation for the electron impact reaction j, and

g) trace species chemistry model for electron-neutral and electron-ion chemistries [6].

We use a two region approximation: a perimeter sheath region (thickness tens of microns) and a quasi-neutral bulk region of dimensions hundreds of mm. The plasma DSMC code is used for the bulk region and the 1D sheath model is used to compute the potential jump from the surface to the bulk. Note that a special treatment is used for modelling the trace, or infrequently occurring species. Since the ionization fraction is very low (~ 0.001), the ionization reaction rates must be low; however, the trace species are the ions which physically drive the entire system response. This model is described in detail in ref. [6].

3 Electron Energy Equation

A critical modelling issue is how to compute the electron energy or temperature; the behavior of processing plasmas is dominated by the plasma chemistry whose rates are a direct function of the electron temperature. Unlike prior DSMC plasma applications for hypersonic driven plasma flows DSMC [7,8,9] where the quasi-neutral electrons are 'attached' to an ion and transported with it, we simply compute the local electron density, n_e, based on the average cell ion density. This *cell centered* electron number density is now used in the ambi-polar electric field model, to determine the chemical reaction rates, and in the energy equation. We have investigated two strategies for determining the electron energy or temperature: 1) invoke a control volume energy balance on the cell (item f above) or 2) periodically during the simulation, we freeze the heavy particle (neutrals and ions) transport and collision, generate kinetic electrons at the local charge density, and take many electron time steps to compute the average steady state cell electron energy. The first strategy assumes a Maxwellian EEDF and solves for an equivalent electron temperature and therefore uses reaction rates; the second

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strategy does not assume an EEDF and uses energy dependent cross-sections to determine reaction probabilities. The first approach is used in the present work since it is more computationally efficient (for design simulations) and the EEDF in a low pressure plasma system is typically Maxwellian since the electron-heavy collision frequency is much lower than the probability of the electrons oscillating between the reactor walls.

4 Implementation Issues

The computation of the electron energy equation, even by the simpler and more efficient control volume formation, is too expensive to perform every time step. Also, since gradients are computed on statistical quantities, using a small, ensemble average can help smooth the fluctuations. However, if the electron temperature is held constant and the electron density, n_e, is computed every time step, an instability will occur. For example, if the electron density increases due to statistical fluctuations while T_e is unchanged, the electron impact chemistry will increase which will result in a spiraling increase of n_e. To mitigate this problem, we hold the local electron energy, $3/2\ n_e kT_e$, a constant between updates of T_e; that is, since we desire the steady state response, we keep the product, $n_e T_e$ a constant between each update of the electron energy equation. For the gedanken problem with the fluctuating increase in n_e, T_e will decrease which will adjust the ion production.

The test case is an electro-negative plasma; that is, the negative ion fraction is comparable to the positive ion fraction and is much harder to simulate than a electro-positive plasma such as Argon. The difficulty is the tight coupling between the plasma chemistry and the system behavior: an attachment reaction produces a negative ion and reduces the electron density which changes the electron impact chemistry which affects everything else! For the bulk plasma region, negative ions are specularly reflected by the sheath (sheath potential >> ion energy) and the flux of the positive ions into the sheath is determined assuming the ions have the Bohm velocity, $V_{Bohm} = \{\ kT_e\ /\ m_i\ \}^{1/2}$ where m_i is the mass of the ion at the sheath edge.

The gradients in the electric field equation and in the electron energy equation are computed using shape functions similar to those in traditional finite element or finite control volume methods. These functions are pre-computed at initialization time and the evaluation is very fast.

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5 Validation: Cl_2 Glow Discharge Plasma

The experimental data used for model comparisons was taken on a Gaseous Elec-
.tronics Conference (GEC) rf Reference Cell which had been modified with an in-
ductively coupled source as shown in Figure 1.[10] The powered source is a five-
turn coil at the top of the reactor, power by an rf power supply at 13.56 MHz. The
gas injection was through a side port of the reactor at a rate of 5 to 20 sccm of
chlorine. The experiments spanned from 20-50 mtorr and 185-300 W of deposit-
ed rf power.

The Icarus grid used to model the GEC reactor is shown in Figure 2. The geome-
try of the reactor was simplified for the simulations, to axisymmetric 2D, by not
including the outer ports. The reduction in surface area at the outer radius was
compensated for by proportionally increasing the wall recombination constants
for surface chemistry. A full 3D simulation is not justified for this system since
the very low neutral flow velocities indicate that this system behaves in a strong
diffusive manner. The wall temperatures in the simulation were 350K.

The chlorine plasma was modeled using five species; Cl_2, Cl, Cl^+, Cl_2^+, and Cl^-.
A description of the gas phase and surface reactions used in the Icarus simulation
are given in Table 1. The electron excitation reactions, needed for the energy
equation, are not included in this table. The electron reactions included the
Frank-Condon effect by adding the additional energy to the heavy products. A
surface recombination probability of 0.1 was used to describe atomic chlorine re-
combining to form molecular chlorine. Positive ions were neutralized at the wall
and reflected with 100% thermal accommodation. The negative ion, Cl^- was re-
pelled from the reactor walls (sheath) due to the magnitude of the sheath poten-
tial.

GEC reactor simulations for a chlorine plasma required approximately 1×10^6
computational particles for a grid with 1,170 computational cells; this included a
trace species ratio of 1-200 for the ions. Icarus simulations were performed on a
1024-node nCUBE-2 massively parallel supercomputer. Typical simulations
required $5\times10^5 - 7\times10^5$ computational time steps with a neutral time step of
1×10^{-6} sec; an ion subcycling factor of 3 was used. Typical simulation times were
8 - 12 hours on 512 nodes. A complete discussion of the comparisons between
Icarus (without the self-consistent energy equation) and the experimental data
can be found in ref. 11. Prior simulations used the local $n_e T_e$ constant from a

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continuum based code [12].

Figure 3 shows the comparison for the measurements of Cl+ metastable ion density and the Icarus predictions for several radial profiles measured from the lower electrode for 185 W of power and 20 mTorr. The experimental LIF measurements only yield a qualitative result; the plots have been normalized to their respective maximum values. The predicted trends are in reasonable agreement Figure 4 shows a countour of the Cl- ion density distribution. The experimental data has strongly indicated a off-centerline peaked concentration. The Icarus simulation is the first simulation to predict this; however, the peak is higher than indicated by the experimental measurements. This can be due to either infrequent updating of the ICP power with the new electron density distribution or that the current simulations do not include a pre-sheath region which would tend to move the Cl- peak away from the wall. The magnitudes of the predictions are within the experimental uncertainty.

6   Conclusion

We have presented a self-consistent plasma DSMC model for simulating chemically reacting plasma systems. The inclusion of a consistent electron energy equation has increased the agreement between the simulations and the data. This first time prediction of the off-centered Cl- ion distribution qualitatively agrees with the data; however, further investigation is needed to improve the quantitative agreement. We believe the differences between prior continuum and the present simulation can be attributed to energy-dependent cross-sections now being used for ion transport and the inclusion of charge exchange reactions which can create 'cold' ions. This bi-modal ion energy distribution is not modelled by a single ambi-polar diffusion coefficient.

We are undecided about the trade-offs between a control-volume versus a quasi-neutral kinetic electron energy equation. We consider this as open issue for these classes of problems, those with mild gradients, at the present time.

7   Acknowledgments

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8 References

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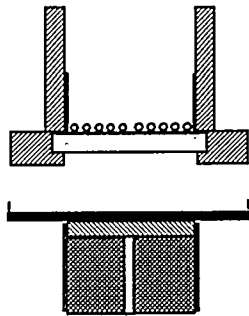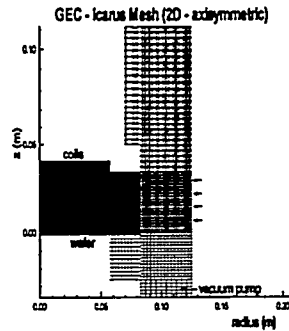
Fig. 1: GEC ICP Configuration

Fig. 2 Icarus Mesh

Table 1: Plasma Chlorine Mechanism for Icarus Simulations

Gas Phase Reactions	Reaction Rate Expressions (m³/s)[a]	Frank-Condon (J)
1.) $Cl_2 + e \rightarrow 2Cl + 2e$	$3.99 \times 10^{-14} \cdot T_e^{0.115} \cdot e^{(-4.43/T_e)}$	0.96×10^{-19}
2.) $Cl_2 + e \rightarrow Cl_2^+ + 2e$	$2.13 \times 10^{-14} \cdot T_e^{0.771} \cdot e^{(-11.7/T_e)}$	
3.) $Cl + e \rightarrow Cl^+ + 2e$	$2.96 \times 10^{-14} \cdot T_e^{0.554} \cdot e^{(-13.1/T_e)}$	
4.) $Cl_2 + e \rightarrow Cl + Cl^-$	$2.21 \times 10^{-16} \cdot T_e^{0.485} \cdot e^{(-0.174 \cdot T_e)}$	5.78×10^{-19} .
5.) $Cl^- + e \rightarrow Cl + 2e$	$2.95 \times 10^{-14} \cdot T_e^{0.6801} \cdot e^{(-3.7994/T_e)}$	
6.) $Cl_2^+ + e \rightarrow 2Cl$	$9.00 \times 10^{-13} \cdot (300./T_e \cdot 8.61753 \times 10^{-5})^{0.61}$	1.84×10^{-18}
7.) $Cl_2^+ + Cl^- \rightarrow 3Cl$	$5. \times 10^{-14}$	
8.) $Cl^+ + Cl^- \rightarrow Cl + Cl$	$5. \times 10^{-14}$	
9.) $Cl_2^+ + Cl_2 \rightarrow Cl_2 + Cl_2^+$	0.75 (probability), $120 \times 10^{-20} (m^2)^b$	
10.) $Cl^+ + Cl \rightarrow Cl + Cl^+$	0.75 (probability), $120 \times 10^{-20} (m^2)^b$	
11.) $Cl^- + Cl \rightarrow Cl + Cl^-$	0.75 (probability), $120 \times 10^{-20} (m^2)^b$	
Surface Reactions		
$2Cl \rightarrow Cl_2$	0.1, reaction probability	

[a] Rate expressions were generated from cross sections assuming a Boltzmann electron energy distribution function for a range of E/N values.
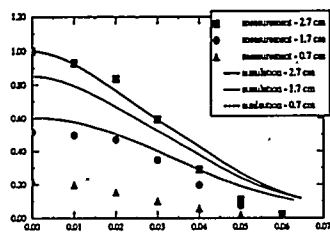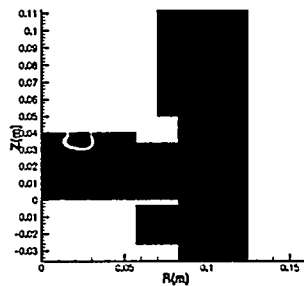[b] Charge exchange reaction probabilities are computed using a specified probability and cross section

Fig. 3 Radial profile comparisons of Cl+

Fig. 4 Contours of Cl- density

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