Simulations of multipactor-assisted breakdown in radio frequency plasmas

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Breakdown in low pressure radio frequency (rf) plasmas is investigated with particle-in-cell simulations in a model gas based on argon. A one dimensional model with a realistic rf wave form is used in a range of pressures between 1 and 50 mTorr. Dynamic scaling and electron generated secondaries are introduced in order to simulate breakdown realistically, and it is possible to follow the discharge development from a few initial electrons to the steady state plasma with \( n_e \sim 10^9 \text{ cm}^{-3} \). The results confirm that breakdown is controlled by the resonant multiplication of secondaries known as the multipactor. For the pressures investigated, however, ionization is the main source of new charges and a model based on a typical electron trajectory is introduced to account for the growth in the number of electrons in the discharge gap. The model is in good agreement with the simulation results. © 1996 American Institute of Physics.

I. INTRODUCTION

It has been known for many years that the initiation of breakdown in low pressure radio frequency (rf) plasmas is controlled by resonant multiplication of secondary electrons at the wall of the discharge vessel.\(^1\)\(^-\)\(^2\) This resonant multiplication, known as the “multipactor” effect\(^3\) is important when the neutral pressure is so low that the mean free path for electron scattering exceeds the length of the discharge gap. Under favorable conditions determined by the values of the interelectrode distance, the rf frequency and amplitude, and the secondary yield coefficient \( \delta \) of the wall material, low energy secondary electrons emitted at one electrode are accelerated by the electric field and reach the opposite electrode just as the rf field goes through zero and reverses sign. These electrons strike the wall at high energy and if \( \delta > 1 \) more secondaries are produced than primaries lost. Multiplication occurs in each half cycle of the rf field and this leads to an exponential growth of the number of electrons in the discharge gap.

In this paper we present results of particle-in-cell (PIC) simulations of breakdown in a parallel plate rf discharge. Multipactor controlled breakdown can be expected to occur under conditions which are typical of some modern plasma processing reactors\(^4\) and our motivation for this work is the increasing importance of time modulation, and in particular pulsing of the discharge in plasma processing.\(^5\) As the pulse time gets shorter, the initial nonequilibrium phase of the discharge starts to dominate and it is essential to model this realistically when simulations of pulsed plasmas are attempted. We are thus mainly concerned with devising simulation tools which can assist in the development of applications for pulsed plasmas.

The details of the simulation technique are given in Section II and the results are presented and discussed in Section III which is followed by a conclusion.

II. SIMULATIONS

The simulations presented in this paper are based on the PIC/MCC (particle-in-cell, Monte Carlo collisions) algorithm which has been used extensively in rf discharge modeling.\(^6\)–\(^9\) This technique is well-suited to breakdown modeling because it naturally handles both the essential time dependence and the highly nonequilibrium (non-Maxwellian) particle distributions which are to be expected when large fields are present in the discharge.

Our code has been developed over a number of years\(^6\)–\(^11\) and it will not be described here in detail since it uses numerical techniques which are standard in bounded PIC simulations.\(^12\)–\(^14\) Briefly, it is one dimensional, electrostatic, and voltage driven. An external RLC circuit is included although the circuit inductance and resistance are set to zero and the external capacitance is held fixed at 100 pF in the present work. Collisions between charged species and the neutral gas are handled by the null collision method.\(^12\) The model gas is based on argon with ionization, excitation and elastic scattering for electrons, and resonant charge-exchange and isotropic scattering for ions all included.

The generation of secondary electrons at the walls is included in the simulation by considering explicitly each electron which arrives at the boundaries of the simulation region. The kinetic energy of the electrons is used to derive a probability of secondary electron emission and realistic energy distributions functions are used for the secondary electrons. Secondary electron emission due to electron impact has been studied for a wide range of surfaces\(^15\)–\(^16\) and our treatment is based on the experimental data. The secondary electron emission coefficient of many materials shows very similar behavior as the energy of the primary electrons is varied. The coefficient \( \delta \) starts near zero at low energies, rises to a maximum value \( \delta_m \) at an energy characteristic of the surface, and then falls gradually. We use a dependence given by

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\[
\frac{\delta}{\delta_m} = e^2 \frac{E_p}{E_m} \exp\left[-2 \left(\frac{E_p}{E_m}\right)^{1/2}\right]
\]

where \(E_p\) and \(E_m\) are the energy of the primary electron and the energy at which the coefficient reaches its maximum value, respectively, and \(e\) is the base of the natural logarithm. This formula is both simple and a good representation of measured yield curves. More realistic expressions including the impact angle dependence of the yield can easily be implemented but such refinements are not expected to significantly affect the present results.

The energy distribution of the secondary electrons is in general quite complex since it contains a number of different electron groups. First, there are the reflected primaries which undergo only elastic scattering near the material surface before being ejected again. Second, there is a group of low energy electrons which are considered to be true secondaries released from the material. Finally there is a third group—far less numerous—of primary electrons which have undergone inelastic scattering within the material and rediffused back to the surface to be ejected again. We model these distributions by separating reflected primaries and true secondaries and ignoring the small group of inelastically scattered primaries. A constant reflection coefficient of 0.2 is used for primary electrons which suffer only a change in direction when they are injected back into the simulation region. True secondaries are taken to have an energy distribution given by a Gaussian flux with an effective temperature of 3 eV and all secondaries are emitted with a cosine angular distribution.

For the purpose of this study we take the maximum yield \(\delta_m = 2.4\) at a primary energy of 400 eV. These values are typical of oxides and glass and are also reasonable for metal surfaces which are either gas covered or partially oxidized. It should be noted that any value of \(\delta_m > 1\) will result in the multipactor for some favorable set of external parameters. Although secondary electrons generated by ion impact can be important in certain types of plasmas, they are unimportant in the present study since there is essentially no ion loss during the breakdown. Ion-produced secondaries are included in the code but since they only produce small changes in the steady state values of the plasma parameters they are turned off in the simulation runs described below.

Three more issues must be dealt with before the results are described: the scaling problems which arise when a particle simulation is started from one or a few electrons and the density develops through many decades of exponential growth in density so this expedient is inapplicable and charge scaling must be made dynamic in these breakdown studies. We have chosen a very simple algorithm for dynamic charge scaling in the simulation. Each particle is assigned a value of \(\xi\) when it is introduced into the simulation, either through an ionizing collision or secondary emission. A maximum number of particles is chosen and when this is reached the value of \(\xi\) for new particles is doubled. At the same time a new maximum number is chosen. Ionizing collisions by all electrons are recorded but new particles are introduced only with a probability of \(\xi_e/\xi_t\) where \(\xi_e\) is the scaling of the ionizing electron and \(\xi_t\) is the current scaling. In order to guard against inappropriate doubling of \(\xi_t\), successive choices of the maximum particle number add terms from the series \(1 + 1/2 + 1/4 + \ldots\) to the first choice \(N_{\text{max}}\) and a time delay of one half of a rf cycle is used to allow some electrons to escape before \(N_{\text{max}}\) is examined again.

The approach just described works very well for keeping electron superparticle numbers within reasonable bounds, but ions—with their much longer residence time—must be treated differently. At any one time the ion superparticle population contains a range of \(\xi\) values. When the ion particle number exceeds a predetermined bound (set somewhat higher than the maximum number of electron particles) the ensemble is examined and the minimum value of \(\xi\) found. Half of the particles with this minimum value are deleted whereas \(\xi\) is doubled for the other half. This procedure results in extra noise in the electric field, but parametric studies show that this is not a significant problem.

The simple external circuit used in the simulations does not represent a laboratory system realistically. We use a fixed blocking capacitor in series with the discharge, but even including a series inductance is not adequate to model a real system in which a resonant matching network with non-negligible loss is used between the rf generator and the powered electrode. The rise time of the rf signal on the driven electrode is limited in practice by the rise time of the rf generator and the Q factor of the resonant matching network. In order to minimize complications introduced with additional external circuit elements, we have chosen to impose a rise time on the rf voltage wave form. The time dependent voltage from the voltage source is given by

\[
V(t) = V_{\text{rf}} \sin(\omega_{\text{rf}} t) \exp(-\tau_r/\tau_t),
\]

where \(V_{\text{rf}}\) and \(\omega_{\text{rf}}\) are the amplitude and angular frequency of the rf signal and \(\tau_r\) and \(\tau_t\) are the rise time and elapsed time, respectively. With \(\tau_t = 500\) ns, this wave form is a realistic model of laboratory measurements.

The multipactor effect cannot begin unless there are suitable free electrons in the discharge gap which can be accelerated to the wall and release secondaries. These free electrons are necessary for breakdown in all plasmas and different mechanisms may produce such electrons; the two most common examples are ionizing radiation such as cosmic rays and field emission at microscopically rough wall regions. It may initially be thought that when the plasma is
pulsed, some electrons remaining from a preceding pulse can start the discharge therefore initiating the simulation by first loading a cold background distribution is viable. This cannot be done for the following reason. As mentioned above, the rf waveform has a finite rise time. At the same time, multiplication has a definite energy threshold [see Eq. (1)], as does ionization. These facts together imply that at low pressure any cold electrons are swept out of the discharge gap without ionizing the neutral gas or producing secondaries. A constant source of free electrons must therefore be present for break-
down to commence and this is introduced by simply loading cold electrons at random times during the rf cycle as the rf amplitude is rising. Once multiplication starts, this loading is stopped.

### III. RESULTS AND DISCUSSION

The plasma reactors used in industrial plasma processing represent an enormous variety of gas mixtures and wall materials. The conditions under which these reactors operate imply that the state of the walls is in most cases poorly characterized and it is not clear how an appropriate secondary emission coefficient should be determined. We have approached this problem by choosing a typical value of $\delta_m = 2.4$ at $E_m = 400 \text{ eV}$ which is used to represent glass or quartz covered surfaces. We note that a high coefficient can be expected in many cases where metal electrodes are covered, or partially covered, by an oxide layer such as is the case of aluminum.

The simplest way to find the conditions where multipactoring operates is to follow the experimental procedure: the rf voltage is slowly increased until exponential growth in the number of electrons is observed. Fitting this growth curve gives the breakdown voltage. In this work we keep the inter-electrode gap fixed at 5 cm and vary the rf frequency, amplitude, and the gas pressure. Breakdown voltages are found by running simulations at very low pressure ($<0.01 \text{ mTorr}$) in order to minimize run times. These “zero pressure” breakdown voltages compare well to both theory and experiments, however, while we have pursued these calculations at some length, the results will not be reported in this paper which deals with neutral pressures more relevant to plasma processing. We note that our simulation technique presents a viable alternative to other methods, and is particularly useful when the long-time development and the influence of space charge are of interest.

The development of the discharge breakdown is divided into two periods which are clearly seen in Fig. 1 where the electron and ion densities are shown with the potential mid-line moving together in the discharge gap. The electrodes are driven by a potential $V$ and by secondary generation and in the electropositive discharge under consideration they will be lost only at the walls. The growth in density can be expected to last many rf cycles and by secondary multiplication and the rise time of the rf voltage is also much greater than a rf period so we separate the time scales and consider the ionization frequency $v_i$ and the secondary multiplication factor $\delta$ averaged over half a rf cycle. To further simplify the problem we consider a single sheet of $N_e$ electrons per unit electrode area moving together in the discharge gap. The number of electrons per unit area $s$ released from the driven electrode at position $s=0$ with a phase $\phi_0$ gives

$$\frac{dN_e}{dt} = \left( \nu_i + \frac{\omega_0(\delta-1)}{\pi} \right) N_e,$$

where the $\omega_0/\pi$ factor implies an average over half a rf cycle and the secondary yield $\delta$ is to be evaluated at the energy corresponding to the impact velocity of the primary electrons.

#### A. Secondary yield

We now consider the motion of an electron between the two electrodes. Following Vaughan we consider one electrode driven by a potential $V_0 \sin(\omega_0 t)$ so integrating the equation of motion for an electron with an initial velocity $v_0$ released from the driven electrode at position $s=0$ with a phase $\phi_0 = \alpha$ gives

$$\frac{ds}{dt} = \frac{eV_0}{m_e \omega_0} (\cos \alpha - \cos \omega_0 t) + v_0$$

![FIG. 2. The applied voltage (dotted) and the components of the electron current at the powered electrode. The primary (positive), secondary (negative), and the total currents are shown.](image)
where $d$ is the distance between the electrodes. The position is given by integrating again

$$s = \frac{eV_{rf}}{m_e\omega_r^2d}[(\omega_r t - \alpha)\cos \alpha - \sin \omega_r t + \sin \alpha] + \frac{v_0}{\omega_r} (\omega_r t - \alpha).$$

(6)

If multiplication is to occur we must have $s=d$ when $\omega_r t = \pi + \alpha$ (only the lowest order multipactor for which the electrons take half a rf cycle to traverse the interelectrode gap is considered). Putting this condition into Eq. (5) the impact energy expressed in electron volts is

$$V_i = \frac{m_e}{2e} \left( \frac{2eV_{rf}}{m_e\omega_r^2d} \cos \alpha + v_0 \right)^2.$$  

(7)

The multiplication factor $\delta$ in Eq. (4) can be found by solving (6) with $s=d$ for the phase at the given applied voltage $V_{rf}$ together with Eq. (7). However, in contrast to the usual analysis of the multipactor effect where the concern is simply the existence of a multipacting discharge (i.e., the minimum or maximum voltage at which it occurs), we are imposing a value of the voltage and frequency and have to consider solutions with $\alpha<0$. In fact with $s=d$, Eq. (6) has two solutions in $[-\pi, \pi]$ and only the negative phase solution corresponds to an arrival energy with a secondary yield $\delta>1$. This implies that many of the secondaries will be lost again since for $\alpha<0$ the electric field still accelerates electrons back to the electrode. Estimation of the actual yield (after the loss of some secondaries back to the wall) is complicated by the fact that the primaries arrive with an energy and phase distribution which is affected by collisions in the discharge gap, but examination of the wall currents in the simulation gives the effective yield directly.

The electron primary, secondary and total currents to the powered electrode are shown for three rf cycles in Fig. 2. The currents are shown for $p=5$ mTorr, during the rise in applied voltage when $V_{rf}=190$ V and the time dependence of the net current shows the production and subsequent loss of the secondaries. As is clear from the figure, the yield is close to 1 and this is confirmed by taking the ratio of total primary and secondary currents in each rf cycle. The effective secondary yield is very weakly dependent on the rf voltage with increasing neutral pressure reducing the yield. This is to be expected since the electrons undergo inelastic collisions in the gap so the average impact energy of the primaries is reduced at higher pressure. The effective yield is shown as a function of pressure in Fig. 3 for $f_{rf}=25$ MHz and $V_{rf}=250$ V. The data points are to be understood as average values over the whole period between the start of the density rise and plasma formation.

**B. Ionization rate**

An estimate of the ionization frequency can be obtained by considering a representative electron trajectory. With regard to the discussion above, it is apparent that we can consider this representative trajectory to start at $\alpha=0$ with $v_0>0$. The half cycle-averaged ionization rate is given by

$$k_i = \frac{1}{\omega_r} \int_{\alpha_1}^{\alpha_2} v \sigma(v) d\alpha'$$

(8)

where the integral is taken over the phase $\alpha' = \omega_r t$ with $\alpha_1$ the phase at which the electron first attains the ionization energy $E_i$ and $\alpha_2$ the phase at which the electron reaches the wall. Using (5) with $v_0=0$ and $\alpha=0$ gives

$$\alpha_1 = \cos^{-1} \left( 1 - \frac{\omega_r d}{V_{rf}} \sqrt{\frac{2E_i m_e}{e}} \right).$$

(9)

The upper limit follows from solving an equation of the form $c\alpha = \beta - \sin(\beta)$. In order to avoid a numerical solution we approximate the right hand side by $\beta^{2/3}/\pi^{1/2}$ which provides an excellent fit in the region of interest and gives

$$\alpha_2 = \pi^{6/11} \left( \frac{d^2 \omega_r^2 m_e}{e V_{rf}} \right)^{2/11}.$$ 

(10)

Finally, the ionization cross section can be approximated by

$$\sigma(E) = q_0 \frac{\ln(E/E_i)}{E E_i}$$

(11)

with $q_0=3.12 \times 10^{-17}$ m$^2$ V$^{-2}$ and $E_i=15.76$ eV in argon.
Fig. 4. Several points are worth noting. To reproduce the lengthy analytic result here and show instead a comparison between the model and the simulation results in Fig. 4, several points are worth noting:

1. Consideration of the representative trajectory provides a good model at a low pressure. As the neutral density rises, the average primary electron energy is affected by energy loss through excitation and ionization collisions resulting in a lower ionization frequency in comparison with the model. At still higher pressures, the electrons created in ionizing collisions also contribute and complicate the estimation of the ionization rate.

2. The arrival time of the primary electrons at the wall agrees well with Eq. (10). For the conditions in Fig. 2, we obtain \( \alpha_2 = 0.730\pi \). The peak in the primary electron current occurs at 0.76\( \pi \).

3. The ionization rate is only weakly dependent on the rf voltage and this can be understood by considering the phase interval \( (\alpha_2 - \alpha_1) \) which decreases as the voltage increases. Although the peak electron energy increases with voltage, this is offset by the shorter phase interval resulting in an almost constant cycle-averaged ionization rate.

4. There is a threshold implied by the phase matching condition for the multipactor. In the context of the present model this can be written \( \alpha_2 = \pi \) which leads to a threshold voltage of 111 V. The density does not begin to rise until this voltage is reached, in good agreement with the data in Fig. 1.

C. Plasma formation

For the conditions chosen for this study, the multiplication of secondary electrons does not lead directly to the observed density rise. Ionization is responsible for the increase in electron number and the number of ions is thus comparable to the number of electrons during the whole development. However, the ionizing collisions are mainly due to accelerated secondaries so the phase matching which is the basis of the multipactor is essential.

During the time when the discharge is dominated by secondary emission it has an interesting structure, consisting of an electron bunch accelerated across the gap and partially neutralized by uniformly distributed cold ions. It is a complex and interesting problem to calculate the effects of space charge in this system, and indeed to define a shielding length, but we may estimate the time of transition to a plasma by working back from the fully formed plasma and using the sheath formation criterion (3) given above. Using only the average energy parallel to the electric field we find that \( \lambda_D = 1 \text{ cm} \) at about 2.75 \( \mu \text{s} \). Since the electron distribution is not Maxwellian (not even isotropic) at this time, this is only a rough estimate, but it is in good agreement with the time when rectification of the applied rf voltage begins [Fig. 1(b)].

The exponential growth model discussed in the previous section gives a good account of the growth in the number of electrons in the discharge gap as this goes through 8 orders of magnitude. After about 2 \( \mu \text{s} \), the density in Fig. 1 shows a gradual decrease in the growth rate before the transition from a multipacting to a fully formed discharge. This is mainly due to a gradual increase in the wall loss rate which is a consequence of space charge spreading the phase distribution of the primary beam. The transition to a plasma is shown in Fig. 5 where the electron density is plotted as a function of position and time. During the secondary sustained growth the electrons are concentrated in a bunch with the maximum density initially at the electrodes. As the electrons are accelerated away from the electrodes by the rf field the density drops since flux is approximately conserved. Once sheaths begin to form, a centrally peaked electron density distribution builds up. As can be seen from the figure, electrons accelerated away from the walls still make a substantial contribution to the ionization rate immediately after the transition. A characteristic phenomenon accompanies this transition: a pulse of high energy electrons lasting several rf cycles reaches the electrode. A similar electron pulse has been observed during breakdown in a low pressure helicon plasma.\(^{21}\)

After the formation of the sheaths the potential between the electrodes [Figure 1(b)] rises above the instantaneous potential at either electrode and eventually approaches
\[ V_p = \frac{V_{rf}}{2} \left[ \sin(\omega_{rf} t) + 1 \right] \]  

as expected for the plasma potential in a capacitive rf discharge. The resulting nonzero average field accelerates ions to the electrodes. Since this period begins with a very high electron temperature, the average plasma potential is initially considerably higher than the steady state value and the first group of ions reaches the electrode with somewhat higher average energy. This phenomenon has also been observed experimentally.\(^{21}\)

The results which we have presented so far clearly indicate the importance of secondary electrons in low pressure breakdown. We wish to emphasize that this fact has been recognized for many years and it is treated in the older publications.\(^1\)\(^\ddagger\)\(^2\)\(^\dagger\)\(^2\)\(^\ddagger\) However, our approach allows us to study the role of secondaries at arbitrary pressure and to investigate the transition from secondary controlled breakdown to diffusion controlled breakdown in detail. This is of particular interest in the design of low pressure rf plasma sources and when pulsing or power modulation is used in applications. By considering the role of the secondaries, it is, for example, possible to minimize the plasma ignition time. At low enough pressure it may well be that the plasma does not ignite at all if the configuration has been designed without consideration of the multipactor and it is a common laboratory observation that a rf plasma can often be maintained at a much lower pressure than that at which it will ignite. Other possible developments of this work include choosing the power modulation function and the electron growth rate so as to maximize the power dissipation in inelastic collisions such as excitation or molecular dissociation without at the same time accelerating ions to high energies in the sheaths. Figure 1 clearly points to the possibility of achieving this.

As an illustration of the results which are readily obtained, we consider a pulse of rf lasting for 4 \(\mu s\) and examine the density at the end of this rf pulse as a function of neutral pressure and excitation frequency. Figure 3 shows the density for 5, 20, and 50 mTorr with \(f_{rf}\) between 10 and 30 MHz. The rise time of the applied voltage is 0.5 \(\mu s\) and \(V_{rf} = 250\) V in all cases. At 5 mTorr there is no breakdown at low frequencies and the threshold is close to the boundary where the zero-pressure multipactor exists. The region in which breakdown occurs expands with increasing pressure until at 50 mTorr a discharge forms in the entire frequency range. At this pressure we are in a transitional regime and the representative trajectory model discussed above is no longer relevant. It is important to note that secondary electrons reduce electron wall loss even in the absence of the multipactor and breakdown occurs only when electron generated secondary electrons are included in the simulations for the entire range of parameters investigated in this paper.

IV. CONCLUSION

Breakdown in capacitive rf plasmas has been studied using PIC/MCC simulations in a model gas based on argon. The simulation parameters have been chosen to be relevant to modern low pressure rf plasma sources used in plasma processing. A one dimensional plane parallel system is modeled with one electrode driven by a rf voltage with a rise time of 0.5 \(\mu s\) in a range of frequencies between 10 and 30 MHz and neutral pressures between 1 and 50 mTorr.

The simulations confirm the importance of the multipactor for breakdown at low pressure and allow for the detailed study of the transition from a discharge which is dominated by secondary emission to a fully formed plasma. For a 5 cm electrode gap with \(V_{rf} = 250\) V and \(P = 5\) mTorr, this transition occurs at about 3 \(\mu s\) after the rf voltage is applied. The transition is accompanied by a characteristic ejection of high energy electrons and ions which has also been observed experimentally.

The importance of secondary processes in low pressure breakdown suggests that these processes should be taken into account explicitly when plasma sources are designed and optimized. It may also be possible to choose discharge conditions and configurations which exploit the high electron energies available during breakdown without dissipating power in unwanted ion acceleration in the plasma sheaths.

18. This approach conserves energy only in the time-averaged sense, but scaling studies confirm that the resulting energy loss is negligible.