Bias formation in a pulsed radiofrequency argon discharge

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A one dimensional (1D) particle-in-cell (PIC) computer simulation has been used in conjunction with a small experimental plasma reactor, to investigate the effects of pulsing on a low pressure, capacitively coupled, rf argon plasma. In particular this article investigates the time-constants involved in the development and evolution of the bias voltage in asymmetric reactor geometry. Surprisingly, the charging time for the blocking capacitor does not occur on electron time scales, but is influenced primarily by the ambipolar drift of ions to the earthed electrode. It is shown that following plasma breakdown there is a net current flow in the system which charges the blocking capacitor in the external matching circuit and produces the bias voltage. Both the PIC simulation and the experimental measurements show that a net current flow is produced by a delay in the onset of the electron current to the earthed electrode, which is correlated to the charging time of the capacitor. From the simulation it can be seen that during this period the plasma potential in the center of the discharge is higher than one would expect, preventing electrons from reaching the earthed electrode. © 1997 American Institute of Physics. S0021-8979(97)01614-9

I. INTRODUCTION

Time modulation of high density plasmas has recently shown great potential for improved material processing applications. Pulsed plasmas are also instrumental in fundamental studies of transient, nonlinear plasma phenomena. Experimental studies of breakdown of an argon discharge have been carried out using pulsed helicon and parallel plate rf sources. Recently particle-in-cell (PIC) simulations have been used to study secondary-emission assisted breakdown in low pressure (1–50 mTorr) parallel plate discharges. In order to control the behavior of pulsed plasmas, it is necessary to fully understand the nonequilibrium processes taking place during both the on and off periods of the pulse. Materials processing applications typically require chemically complex plasmas, with many species present, and complicated reactor geometries. This produces a large number of competing physio-chemical effects making results correspondingly difficult to interpret. For clarity therefore, this study confines itself to an argon plasma in a simple parallel plate geometry, and only examines the first few microseconds after the pulse is initiated.

Parallel plate rf discharges have a long history in the materials processing industry, but much of their behavior is still only poorly understood, particularly the time-dependent processes taking place while the plasma is breaking down, and in its approach to equilibrium. There are several important time scales involved, including the duration of breakdown, development of the bias voltage, and evolution of plasma parameters, such as charged particle densities, current, and power to steady state. These time scales can become particularly significant when pulse lengths of the same order (micro- to milliseconds) are used.

This work uses both experiment and computer simulation to study a low pressure, rf, argon discharge in asymmetric reactor geometry, during and immediately after breakdown. Results are presented for the first few microseconds after the pulse is applied, which covers the time of the transition from a “symmetric” discharge, in which the applied voltage is divided equally between the sheaths at the powered and earthed electrodes, to an “asymmetric” discharge in which most of the voltage is dropped at the powered electrode. This transition takes place as the blocking capacitor charges and a bias voltage develops at the powered electrode.

II. EXPERIMENTAL SETUP

The experimental system consisted of a small parallel plate reactor with a 13.56 MHz radiofrequency power source (ENI), modified to obtain rise times on the order of 100 ns of nanoseconds, which is controlled by a pulse generator. This is shown schematically in Fig. 1 in comparison with the simulation configuration. The power was coupled to the live electrode via a L matching network with a 50 dB coupler inserted between the rf generator and the matching network. An rf probe was used to measure the voltages in the matching box and on the live electrode. Live and earthed electrodes were of a similar diameter (~10 cm) and could both be moved to adjust the inter-electrode distance L. The vacuum vessel containing the two electrodes consisted of an aluminium cylinder 30 cm in diameter and 30 cm long, pumped down to about 10^-6 Torr by a turbo molecular pump placed on top of the chamber. Argon gas was flowed continuously through the system with the gas pressure controlled by a flow meter. Axial and radial ports were available for

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that PIC codes self-consistently model the plasma, by calculating the charged particle motion due to applied and internal fields, so that parameters such as the particle energy distributions and density and potential profiles are a product of the plasma conditions, rather than being imposed externally.

The simulation is voltage-driven and includes an external circuit capacitor\(^{14}\) which couples the inner electrode to the generator, while the outer electrode is earthed. The capacitor in the simulation models the blocking capacitor in the experimental matching network, but the rest of the match box is not explicitly represented. As a consequence the rise-time of the voltage signal, which depends on the matching network and response time of the rf generator, must be included empirically in the simulation. For these simulations the source voltage is given an exponential rise-time of the form

\[ V_{rf}(t) = V_{rf} e^{-\frac{t}{\tau}} \sin(2\pi f_r t), \]

where \( V_{rf} \) is the voltage amplitude, \( f_r \) the frequency, and \( \tau \) the exponential time constant.

The simulation models collisions between charged particles and neutral atoms realistically, using analytic fits to real argon cross sections. The null collision method\(^{12}\) is used with isotropic scattering of the particles. Electrons can make ionization, excitation, and elastic collisions, and ions make charge exchange and elastic scattering collisions. Secondary emission and electron reflection from the walls of the reactor are also included.

The spherical electrode simulation was chosen as the simplest method of modeling a system with unequal area electrodes. As the simulation has only one spatial dimension it cannot be expected to exactly model the experimental system, which is intrinsically multi-dimensional. However, the goal of this research is not to model the experiment in exact detail (at this stage), but instead to use the computer code to qualitatively model the physical processes occurring during breakdown in an asymmetric reactor. Relative agreement between the simulation and available experimental results was used to establish the validity of the general behavior of the simulation at this level. Experimental measurements are difficult to perform and interpret in these systems and so the simulation was then applied to performing a more detailed study of the fundamental properties of the system.

### III. SIMULATION

The simulation uses a one-dimensional, electrostatic PIC code, with Monte Carlo collisions, to model a reactor with concentric spherical electrodes. PIC modeling techniques have been well documented in previous publications\(^{10–13}\) and so only a brief description of the code is given here. Note

![Diagram](image.png)

FIG. 1. Schematic showing (a) experimental and (b) simulation configurations.

### TABLE I. Running conditions used for experiment and simulation

<table>
<thead>
<tr>
<th></th>
<th>Experiment</th>
<th>Simulation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Distance (cm)</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>Area ratio: (A_{\text{elec}}/A_{\text{pwr}})</td>
<td>3–15</td>
<td>9</td>
</tr>
<tr>
<td>Pressure (mTorr)</td>
<td>50</td>
<td>50</td>
</tr>
<tr>
<td>(V_{\text{elec}}) P-P (V)</td>
<td>1000</td>
<td>1000</td>
</tr>
<tr>
<td>Rise time (\tau) ((\mu)s)</td>
<td>0.2</td>
<td>0.2</td>
</tr>
<tr>
<td>rf frequency (f_r) (MHz)</td>
<td>13.56</td>
<td>13.6</td>
</tr>
<tr>
<td>Capacitor (pF)</td>
<td>10–100</td>
<td>40</td>
</tr>
</tbody>
</table>

### IV. THEORY

rf systems have previously been modeled using equivalent circuit theory\(^{15–17}\) in which the impedance of the plasma is represented as circuit elements. The plasma impedance in low pressure, low density, rf systems is dominantly capacitive, due to sheath effects. The sheaths prevent electrons reaching the electrodes except for the brief period during the cycle when the sheath collapses, allowing sufficient electrons to escape to balance the ion losses. The ion current is small and essentially constant throughout the cycle. The conduction current in the sheaths is therefore insufficient to carry the rf current, instead current continuity is maintained by the displacement current, which is driven by the large, changing...
electric fields in the sheath. To a first approximation therefore the coupling across the sheath regions in these plasmas is capacitive.

The sheaths act like two capacitors in series and so the voltage division between the sheaths, neglecting any resistive or inductive effects, is given by:

\[
\frac{V_{\text{live}}}{V_{\text{earth}}} = \frac{C_{\text{earth}}}{C_{\text{live}}} = \frac{A_{\text{earth}}S_{\text{live}}}{A_{\text{live}}S_{\text{earth}}},
\]

where \(V\) is the amplitude of the rf voltage dropped across the sheaths at the respective electrodes, \(A\) is the electrode area, and \(S\) the maximum sheath width. Equation (1) indicates that for systems in which the area of the powered electrode is smaller than that of the earthed electrode, more voltage will be dropped across the powered electrode sheath, if the sheath widths are equal. In fact the sheath at the powered electrode is typically larger than the sheath at the earthed electrode and so the effect is amplified. Hence asymmetric reactor geometries produce an asymmetric voltage distribution across the plasma.

The presence of the blocking capacitor in the external circuit decouples the electrode potentials and allows different voltages to be dropped across each sheath. In the current configuration the capacitor charges to produce a negative bias voltage at the smaller, powered electrode. In this article we are concerned with the timescales involved in charging the blocking capacitor once the plasma has broken down, and the consequent transition from a symmetric voltage distribution, when the capacitor is uncharged, to an asymmetric distribution, when the capacitor is fully charged. Pulsed plasma conditions are used so that low charged particle densities exist prior to the voltage turning on, and the blocking capacitor is uncharged. The transition from a symmetric to an asymmetric discharge depends on a number of factors including the risetime of the rf signal on the live electrode, the background gas pressure, and the system geometry.

V. RESULTS

A. Electrode voltage

The evolution of the voltage on the powered electrode, obtained from both experiment and simulation, is shown in Fig. 2. The bias voltage, which corresponds to the average of the rf voltage, is superimposed on both plots. The voltage envelope has been dubbed the “Bird’s head” by Booth et al.\(^8\) in reference to the distinctive form induced by the development of the bias voltage (this can be seen more clearly at slower risetimes). At \(\sim 0.5\ \mu s\) after the rf signal is applied there is a discontinuity in the voltage wave form (more easily visible in the simulation results), which marks the end of the breakdown phase. This corresponds to the time at which the Debye length becomes less than half the inter-electrode gap, allowing the formation of sheaths at the electrodes. Prior to this time the charged particle density in the system is so low that the Debye length is much larger than the system length and sheaths cannot exist. As electrons are heated by the applied voltage and start to ionize, the density of charged species increases, and consequently the Debye length decreases (more detail on the breakdown is given in Vender et al.\(^9\)). The sheaths form very rapidly, producing a marked change in the plasma impedance and causing discontinuity in the electrode voltage.

During the breakdown there is no bias voltage and so the voltage division between the sheaths is symmetric. After breakdown the blocking capacitor starts to charge and the bias voltage [the dotted line in Figs. 2(a) and 2(b)] evolves, resulting in an asymmetric voltage distribution. The bias voltage reaches its steady state value at \(\sim 3.5\ \mu s\) for the experiment and \(5.5\ \mu s\) for the simulation. At \(3.5\ \mu s\) the maximum positive excursion of the powered electrode voltage in the experiment is about \(40\ V\). In a continuous plasma an average steady state plasma potential of \(15–18\ V\) was measured using a retarding field energy analyser (RFEA), which is consistent with an average plasma potential of approximately half the positive excursion of the voltage on the powered electrode. Note that in the experiment the negative excursion of the rf envelope develops much more rapidly after breakdown than for the simulation. This appears to be associated with an impedance resonance occurring between the matching network and the plasma as the sheaths develop, allowing more voltage to be dropped across the plasma. This feature is quite reproducible and occurs when the matching network is tuned to give the best power transfer at the end of the pulse. Similar voltage wave forms have been measured in other pulsed rf systems.\(^8\)\(^,\)\(^18\) The simulation circuit, having no inductive component, cannot produce these resonances.

At first glance it is curious that the bias voltage evolves so slowly, since simplistically it could be expected that the
blocking capacitor would charge very rapidly, due to the flux of highly mobile electrons to the powered electrode. However, the reason for this is obvious when the current flow to the electrodes is considered.

### B. Ion and electron currents

A Langmuir probe was used to measure the ion and electron fluxes at various positions along the main axis between the electrodes. The ions were collected at −75 V and the electrons at +30 V. Experimental results for the fluxes next to the earthed electrode are shown in Fig. 3(a) and the simulated currents to the electrode are plotted in Fig. 3(b). The ions start to arrive at the electrode ∼0.5 μs after the rf pulse is initiated, at the end of the breakdown phase, while both experiment and simulation observe a delay of ∼3.5 μs in the arrival of the continuous electron flux. Experimentally the general form of the fluxes, and in particular the delay in the electron flux, was measured for all positions between the electrodes, up to 1 cm from the live electrode and showed the same evolution.

The simulation shows a spike in the electron current just prior to breakdown. This occurs over several rf periods, once the discharge has reached relatively large densities, but before a sheath has formed to prevent electrons reaching the electrode. This characteristic has also been clearly observed in the experimental results under different conditions, but for this set of running parameters it is masked by high frequency noise.

### C. Plasma potential

With the present system it was not possible to measure the experimental plasma potential, but simulation results of the time-dependant behavior of the potential in the center of the plasma are plotted in Fig. 4. This plot shows that the potential is symmetric about zero during the breakdown phase (when the fields are able to completely penetrate the plasma) until the end of the breakdown, when it develops a sudden and dramatic positive dc offset. During the period when the capacitor is charging the potential in the plasma is always much larger than zero, but over the following 3–4 μs the potential gradually decreases, until at 6 μs the average potential is 70 V. This is considerably higher than the steady state value measured experimentally, but the simulation value is still decreasing at this time.

### VI. DISCUSSION

The ions do not start arriving at either electrode until after the plasma breaks down. This is due to their high inertia in the rapidly oscillating fields of the pre-breakdown plasma—until the sheath and presheath regions have formed there is no average potential gradient in the plasma to which they can respond. The reason for the delay in the onset of electron current to the earthed electrode is not so clear, although it is obviously connected to the charging time of the blocking capacitor. Although it was not possible to measure the currents at the powered electrode experimentally, the simulation results show that there is an effective negative current at the powered electrode concurrent with the positive current to the earthed electrode. In other words, a net current flows in the system for ∼3 μs after breakdown, charging the blocking capacitor and producing the bias voltage on the powered electrode.

To check that this net current flow in the simulation is indeed directly involved in producing the bias voltage, the total charge appearing at the powered electrode was determined by integrating the total current arriving at that elec-

![FIG. 3. Ion (solid line) and electron (dotted line) losses to the earthed electrode for (a) the experiment and (b) the simulation. Note that the experiment measures the fluxes next to the electrode using a Langmuir probe biased to −75 V for the ions and +30 V for the electrons, while the simulation determines the actual currents at the electrode.](image)

![FIG. 4. Evolution of the potential in the centre of the plasma obtained from the simulation.](image)
trode. The evolution of the charge is plotted in Fig. 5. At 3.5 μs the net charge is \(-1.4 \times 10^{-8}\) C, which for a capacitance of 40 pF, implies a voltage of \(-350\) V. This corresponds very closely to the simulation bias voltage measured at that time [see Fig. 2(b)].

The mechanism by which electrons are prevented from reaching the earthed electrode can be determined by considering the behavior of the plasma potential. In the last few rf periods before breakdown a large number of electrons are lost from the plasma, with those reaching the live electrode contributing to charging the capacitor. As mentioned previously, ions have a high inertia during the breakdown phase and therefore no ions can reach the walls. Hence, the large number of escaping electrons produces a very large positive jump in the plasma potential, which consequently prevents electrons from reaching the earthed electrode (see Fig. 4).

After breakdown therefore, there is an ion flux to both electrodes, while electrons can only escape at the powered electrode once each rf cycle, during the brief period when the electrode voltage approaches the plasma potential allowing the sheath to collapse. Due to their greater mobility, more electrons can escape each cycle than ions at the powered electrode, however the net negative flux to the powered electrode must be balanced by the ion flux to the earthed electrode in order to maintain the charge balance in the plasma. A net current is therefore allowed to flow in the circuit, which charges the circuit capacitor (n.b. in the experimental system the effective capacitance between source and ground includes the shunt capacitor and any parasitic capacitances, as well as the blocking capacitor). As the capacitor charges and the bias voltage on the powered electrode evolves, the average plasma potential decreases until it approaches a value at which electrons are no longer prevented from reaching the earthed electrode. The charging of the blocking capacitor occurs at a rate which is essentially determined by the loss of ions to the earthed electrode and is therefore dictated by the diffusion time of ions to this electrode.

Although the simulation and experiment show extremely good agreement in their general behavior, there are some details in which they differ. The most noticeable of these is the rate of evolution of the bias voltage. In the experiment the capacitor charges relatively rapidly and the bias voltage reaches its steady state value within a couple of microseconds. The simulation on the other hand has a noticeably slower charging rate and the bias voltage requires \(-5\) μs to reach steady state. In part this has to do with the system geometry, which controls the fluxes of charged particles to the electrodes. As previously mentioned, the simulation configuration is more restricted than the experiment, and for the chosen area ratio the simulation volume can be substantially different from that of the experiment, particularly since the plasma is not strictly confined between the electrodes. The experimental matching network provides added complications, since it is difficult to exactly determine the effective capacitance between the reactor and the source. Preliminary results also indicate that impedance resonances between the circuit and the plasma play a significant role during breakdown, in particular producing large oscillations in the electrode voltage which appear to significantly increase the electron heating, affecting the charging time. This will be pursued further in later work.

VII. CONCLUSION

During breakdown the discharge is symmetric and the density is too low to support sheath formation, and so ion losses to the surfaces are very small. At the end of the breakdown a large loss of electrons produces a jump in the plasma potential, which then prevents electrons from reaching the earthed electrode. This allows a net current to flow in the system, which charges the blocking capacitor and produces a bias voltage on the live electrode. The charging rate of the blocking capacitor is determined by the diffusion time of the ions to the earthed electrode which for the current set of running conditions, is on the order of several microseconds.