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Original Publication Citation

Stark, R. H., & Schoenbach, K. H. (1999). Direct current high-pressure glow discharges. *Journal of Applied Physics, 85*(4), 2075-2080. doi:10.1063/1.369505

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Direct current high-pressure glow discharges

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(Received 22 July 1998; accepted for publication 13 November 1998)

Stabilization and control of a high-pressure glow discharge by means of a microhollow cathode discharge has been demonstrated. The microhollow cathode discharge, which is sustained between two closely spaced electrodes with openings of approximately 100 μm diam, serves as plasma cathode for the high-pressure glow. Small variations in the microhollow cathode discharge voltage generate large variations in the microhollow cathode discharge current and consequently in the glow discharge current. In this mode of operation the electrical characteristic of this system of coupled discharges resembles that of a vacuum triode. Using the microhollow cathode discharge as plasma cathode it was possible to generate stable, direct current discharges in argon up to atmospheric pressure, with estimated electron densities in the range from 10^{11} to $10^{12}\,\mathrm{cm}^{-3}$. The recently demonstrated parallel operation of these discharges indicates the potential of this technique for the generation of large volume plasmas at high gas pressure through superposition of individual glow discharges. © *1999 American Institute of Physics.* [S0021-8979(99)07304-1]

I. INTRODUCTION

Research on high pressure glow discharges is motivated by applications such as instantly activated reflectors and absorbers for electromagnetic radiation, surface treatment, thin film deposition, remediation and detoxification of gaseous pollution, and gas lasers. The two basic methods which can be used to generate large volumes of weakly ionized gas at high (atmospheric) pressure are: (1) external ionization (by means of photons or charged particles); and (2) internal ionization, the generation of electrons and ions in a self-sustained gas discharge.

Generally the efficiency of external ionization is rather low, and therefore the cost of such a method relatively high. Methods to generate large volumes of ionized gas at high pressure have therefore concentrated on ionization mechanisms where the energy required for the ionization is drawn from electrical energy. Examples of this kind of mechanism are radio frequency (rf) and microwave discharges, barrier discharges, and pulsed corona discharges where the discharge is sustained by alternating or pulsed fields, and steady state discharges where the discharge is driven by a direct current (dc) power source.

Much of the efforts in generating stable glow discharges at high pressure have focused on preventing the onset of instabilities in the regions near the electrodes,[1] particularly in the cathode region. These are the regions of higher electric field and consequently higher power density compared to the positive column of the discharge. This region is therefore the cradle of instabilities which lead to constrictions and arc formation in the discharge.

The glow-to-arc transition (GAT), the development of a highly conductive channel which shorts out the glow discharge, shows the first visible evidence near the cathode.[2] Other instabilities which may develop in the positive column of discharges in electronegative gases, such as the attachment instability, are generally more benign than the GAT.

Segmentation of the cathode, and ballasting the individual discharge resistively has been used to prevent the onset of the GAT instability in atmospheric pressure glow discharges.[1] The current density in the bulk of the glow discharge is known to increase linearly with pressure ($j_{\mathrm{bulk}} \propto p$), whereas the current density in the cathode layer for normal mode operation increases quadratically with pressure ($j_{\mathrm{cl}} \propto p^2$). In order to make the conditions in the bulk and at the cathode compatible, the current cross section at the cathode needs to be reduced with increasing pressure. This was achieved by using pins as individual cathodes with cross sections small compared to the area of the cathode segment.[3] The onset of bulk instabilities can be prevented by flowing air with such a speed through the discharge that the plasma is replaced by cold air on a time scale small compared to the inverse of the growth rate of bulk instabilities.

Another way of eliminating the conditions for GAT in the cathode fall region of a glow discharge is to eliminate the cathode fall, that means to provide the electrons rather than through ion impact at the cathode, through external sources. This requires replacing the cathode by an externally controlled electron emitter. Besides of offering the possibility to adjust the electron emission to the large volume glow discharge, this approach also keeps the thermal losses in the cathode, which in case of resistive ballast are substantial, at a minimum and reduces the requirements for cooling. Experimental studies on the use of microhollow cathode discharges as plasma cathodes have been performed in argon and the results are reported in this article.

II. CONCEPT OF MICROHOLLOW CATHODE SUSTAINED GLOW DISCHARGES

Hollow cathode discharges are glow discharges between a cathode, which contain some kind of a hollow and an arbitrarily shaped anode. Similarity laws[4] indicate that the dis-

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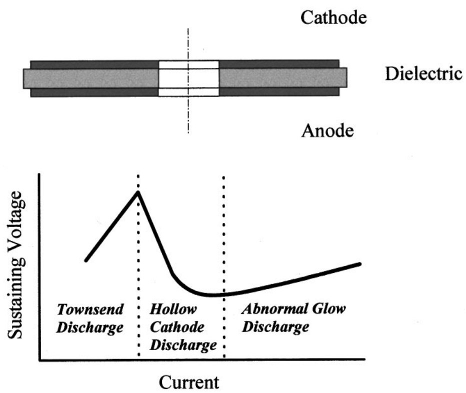

FIG. 1. (top): Geometry of microhollow electrode system with a center borehole. (bottom): Voltage–current characteristic of microhollow electrode discharges (current scale is logarithmic).

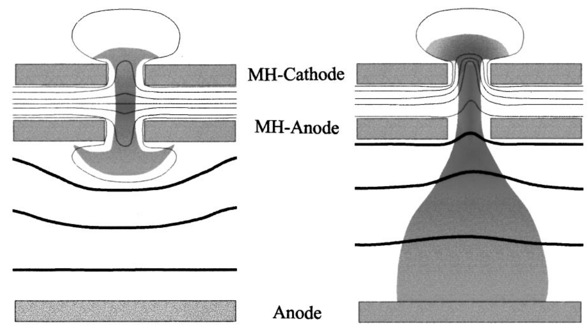

FIG. 2. (left part): Sketch of the microhollow cathode discharge in the Townsend mode and potential distribution in the three-electrode system. (right part): Sketch of the microhollow cathode discharge sustained glow discharge with the MHCD operating in the hollow cathode discharge mode. Also shown is the potential distribution. Because of the change in the potential distribution, the electric field generated by the third electrode becomes comparable to the electric field of the MHCD in the anode aperture and a discharge develops in the space between the MHCD and the third electrode.

charge voltage is constant for constant $p \cdot D$, where p is the pressure of the fill gas and D the diameter of the cathode hole. Experiments in argon and xenon have shown that dc operation of hollow cathode discharges in noble gases is possible at atmospheric pressure, if the diameter of the cathode hole is reduced to values on the order of 100 μm.[5,6] Recent results indicate that stable discharge operation can also be obtained in atmospheric pressure air.[7] The geometry of the microdischarge system is shown in Fig. 1 (top). The electrode system consists of two plane-parallel electrodes with a center borehole in each electrode. As spacer between the electrodes, mica with a thickness of approximately 200 μm is used. The electrode aperture is on the order of 100 μm in diameter. A molybdenum foil of 100 μm thickness is used as electrode material. The currents drawn by a single discharge have for our electrode system been limited to 7 mA due to thermal loading. This value corresponds to an average current density of 120 A/cm^2 in the 100-μm-diam cathode hole area.

The voltage–current (V–I) characteristic of hollow cathode discharges shows three distinct ranges of operation [Fig. 1 (bottom)]. The resistive V–I characteristic at low current, with an exponential increase in current with voltage, indicates that the discharge in this mode is a Townsend discharge.[8] A schematic sketch of the discharge in this mode is shown in Fig. 2 (left part). Here it is assumed that the product of pressure, p, times the electrode gap, d, is less than the $p \cdot d$ value in the minimum of the Paschen curve. The discharge therefore develops along a path, from the outer face of one electrode to the outer face of the second one, rather than the shortest possible path, along the dielectric. At higher pressure, or larger gap between the electrodes, respectively, where this condition is not satisfied, the discharge develops inside the electrode cavity and assumes a hollow cylindrical shape.[9]

With increasing current, the conductivity of the discharge column inside the electrode cavity increases and it forms a virtual anode [Fig. 2 (right part)]. The electric field begins to change from a mainly axial to a more radial field concentrated at the cathode (cathode fall). The axial field is reduced to values required to compensate for electron losses in the virtual anode (positive column). The formation of this strong radial field at the cathode perimeter causes a fraction of electrons generated at the cathode through ion impact to gain such energy that they oscillate through the axis region, unloading much of their energy through ionizing collisions in this region. This hollow cathode effect leads to an increase in current with simultaneous decay in voltage [negative differential conductivity, Fig. 1 (bottom)]. With further increase in current, the normal hollow cathode glow discharge expands over an increasing area at the cathode surface. However, since discharge expansion to areas beyond the circumference of the cathode hole is related to a lengthening of the discharge path, the discharge voltage rises (abnormal glow discharge). This effect which is shown in the V–I characteristics of the discharges at high current [Fig. 1 (bottom)], was also obtained in modeling results.[10]

Extraction of electrons from the microhollow cathode discharge (MHCD) by means of a third, positively biased electrode on the anode side of the MHCD geometry requires that the electric field generated by the third electrode is on the same order as the field in the MHCD. When operated in the Townsend mode, where typical electric fields in the hollow cathode structure are on the order of 10 kV/cm, this would require very high voltages applied to this third electrode which is placed at distance large compared to the gap of the MHCD. Therefore the third electrode, if biased at a moderate voltage, is not expected to have any influence on its operation [Fig. 2 (left part)].

However, when the hollow cathode discharge transfers in the mode where the axial electric field is replaced by a radial one, the electric field generated by the third electrode only needs to be on the order of that in a positive column, typically 100 V/cm to affect the hollow cathode discharge. The potential in the hollow anode plane is then similar to that of an electron lens. The electrons in the hollow electrode rather than drifting to the microhollow anode are rerouted to the third electrode. Consequently it is expected that the MHCD acts as an electron source for a larger volume glow

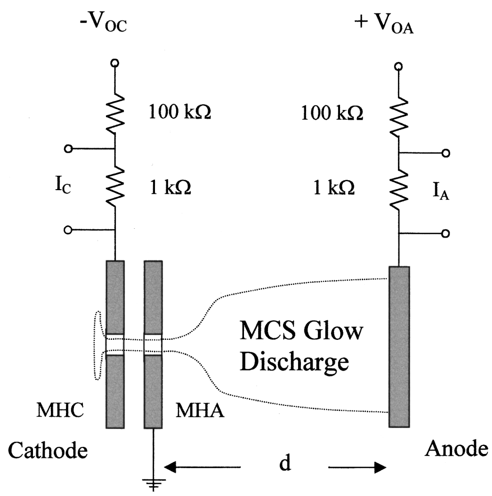

FIG. 3. Electrode configuration and electrical circuit.

discharge between the hollow anode and the third electrode when operated in the hollow cathode mode.

In the resistive current ranges (ranges where the slope of the $V-I$ characteristic is positive) hollow electrode discharges can be operated in parallel without or at least with small ballast. For being used as electron source, as discussed in the previous paragraph, the relevant range of operation would be that at high current, where the discharge resistance increases [abnormal glow discharge, Fig. 1 (bottom)]. Experimental results[11] indicate that parallel operation of microhollow cathode discharges can be achieved in this range of current densities in excess of 100 A/cm^2.

III. EXPERIMENTAL SETUP

The electrode system, which is placed in a stainless steel chamber, consists of a microhollow electrode system (see Fig. 1) and an additional (third) electrode with variable distance from the microhollow electrodes. The vacuum and gas handling system allows us to operate the discharge in the range from millitorr up to atmospheric pressure. Two windows provide optical access to the discharge, side on and end on. A charge coupled device camera with video recording system is used to record the appearance of the discharge.

The electrode configuration and electrical arrangement of the setup is shown in Fig. 3. The electrode on the very left serves as microhollow cathode (MHC) for the MHCD. The anode of the microhollow cathode geometry is on ground potential. The plasma cathode current is measured by recording the voltage across a 1 kΩ resistor in the microhollow cathode discharge circuit. The third electrode, also made of molybdenum, is positively biased. It serves as anode for the microhollow cathode sustained (MCS) glow discharge. Its distance from the microhollow anode, d, can be varied between 0 and 10 mm. As for the MHCD, a 100 kΩ resistor is used to limit the discharge current in the MCS glow discharge, which is measured by means of a resistor of 1 kΩ. The applied voltages at both, the microhollow cathode and the third electrode are limited to values of 800 V, determined by the voltage feed throughs in our discharge chamber. The sustaining voltage of the microhollow cathode discharge is in the range from 100 to 400 V depending on current and gas pressure. The microhollow cathode discharge current was limited to 7 mA to prevent overheating of the sample.

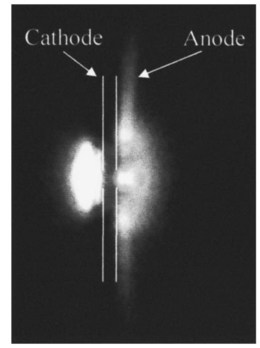

FIG. 4. Side-on view of the microhollow cathode discharge in argon at 160 Torr with the third electrode unbiased.

The discharges were operated in the dc mode. Since possible discharge instabilities may occur on a time scale of microseconds and less, too fast to be observed with dc current and voltage monitors, we have, in all measurements, monitored the current and voltage by means of fast electrical probes and recorded the traces on a 400 MHz digital oscilloscope.

Before each experiment the chamber was evacuated to 10^{-4} Torr, then filled with argon (spectroscopic grade with impurities less than 0.000 01%) at the desired pressure. Argon was chosen because in previous experiments stable microhollow cathode discharges in this gas have been obtained up to atmospheric pressure.[9] The argon pressure in this experiment was varied between 40 and 760 Torr. Two procedures have been used to study the effect of a MHCD on a MCS glow discharge:

At constant pressure and electrode geometry, (1) the bias potential was kept constant and the MHCD current was varied slowly by increasing or decreasing the sustaining voltage, and (2) the MHCD current was kept constant and the potential at the third electrode was varied slowly by increasing or decreasing the bias voltage.

IV. EXPERIMENTAL RESULTS

With the third electrode unbiased the MHCD may develop (statistically) in one of the two modes: (a) an umbrella shaped plasma layer develops at the anode side of the MHCD system; and (b) the MHCD plasma does not extend into the electrode space; only a plasma layer at the circumference of the anode aperture is visible.

An example of the anode plasma extended into the anode backspace is shown in Fig. 4. The side-on photograph shows three areas of high intensity. The area of highest intensity in the center represents the plasma column, the outer areas the edges of the plasma layer at the anode surface. Although the mechanism of the MHCD formation is not yet understood, it will be shown that it is irrelevant for the use of the MHCD as plasma cathode, and we have consequently not paid much attention to this phase.

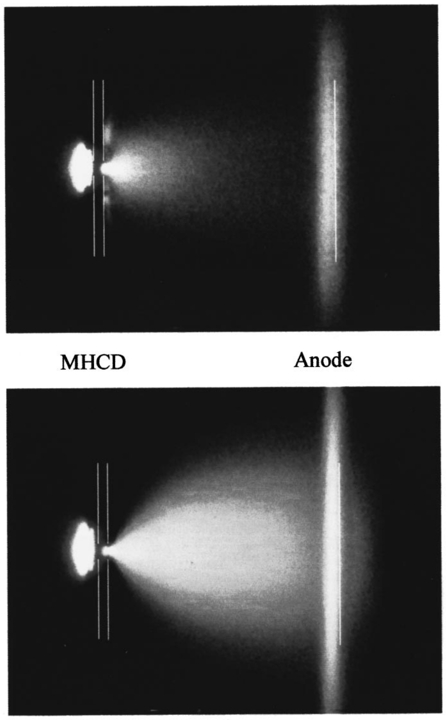

FIG. 5. (top): Side-on view of the MHCD and the predischarge in argon at 160 Torr at a voltage of 66 V applied to the third electrode. (V_{MHCD} $=289$ V, $I_{MHCD}=1.09$ mA, $I_{MCS}=0.42$ mA.) (bottom): Side-on view of the MHCD and the MCS glow discharge for 77 V anode potential. (V_{MHCD} $=259$ V, $I_{MHCD}=1.27$ mA, $I_{MCS}=1.27$ mA.) The gap between plasma cathode and anode is 0.5 cm.

With increasing bias potential, V_A, at the third electrode (anode), the current flow to this electrode increases exponentially, but is still small compared to the MHCD current. In this phase, the *predischarge* phase, a luminous plasma develops in the space between plasma cathode and anode [Fig. 5 (top)]. Although the umbrella like structure of the MHCD anode is still present in this mode, electrons originating from the center of the MHCD are carried increasingly—with increasing V_A—to the third electrode. Eventually, at a certain threshold voltage the plasma umbrella becomes detached from the MHCD anode [Fig. 5 (bottom)] and a bell shaped discharge column is formed. In this mode, the MCS *glow* mode, the current in the MCS glow is identical with the MHCD current.

Besides the dependence on the applied voltage, V_A, the appearance of the glow between MHCD and anode is also determined by the microhollow cathode discharge current. This is shown in Fig. 6. The solid curve in Fig. 6 correspond to the threshold values of MHCD current and anode potential where the transition from the predischarge to the MCS glow is observed. The dashed *vertical* line in Fig. 6 represents the mode of operation where the glow between plasma cathode and anode is controlled by V_A, at constant MHCD current. The dashed *horizontal* line in Fig. 6 represents a mode of operation where the MCS glow discharge is controlled by the current in the microhollow cathode discharge, with the anode voltage, V_A, kept constant. Point A represents the transition from the predischarge to the MCS glow discharge for both cases.

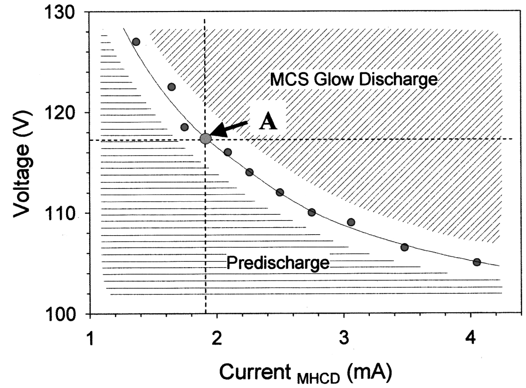

FIG. 6. Range of operation of the predischarge and the MCS glow discharge in argon at 160 Torr.

The development of the current in the glow between plasma cathode and third electrode (anode) with increasing microhollow cathode current (along a horizontal line as shown in Fig. 6: $V_A=$constant) and the corresponding $V-I$ characteristic of the MHCD are shown in Fig. 7. Up to a MHCD current of 3 mA the current in the glow discharge is small compared to the MHCD current. This is the predischarge phase, where the axial MHC electric field in the microhollow geometry exceeds the external field. At the current threshold value in the two fields, the internal and the external fields become comparable and the current is completely rerouted from the microhollow anode to the third electrode.

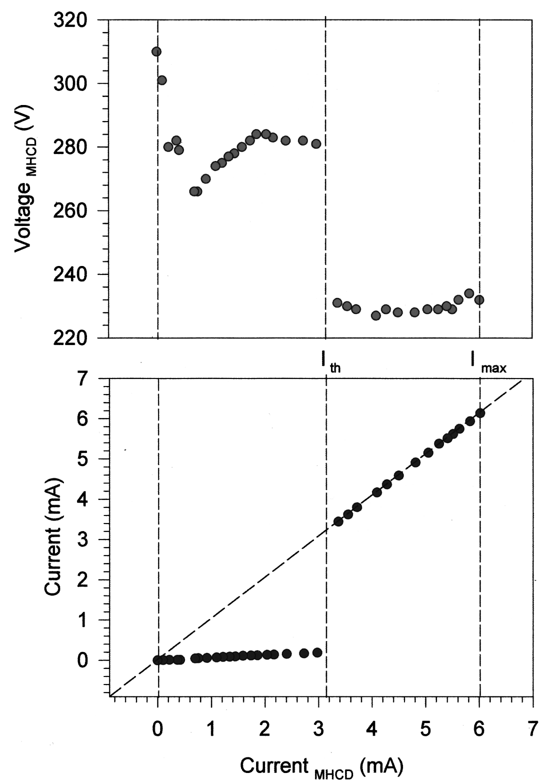

FIG. 7. MHCD voltage (top) and current measured at the anode (bottom) vs MHCD current at a constant anode potential of 100 V (argon, p $=160$ Torr). The transition at I_{th} corresponds to a point on the solid curve in Fig. 6.

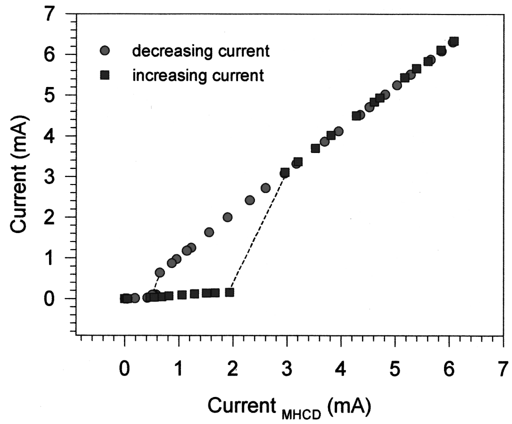

FIG. 8. Hysteresis in the transition from predischarge to MCS glow discharge.

This means, that the MHCD current and the MCS glow discharge current become identical. This switching effect is correlated with a sudden drop in the MHCD voltage. The results depicted in Fig. 7 were obtained by varying the MHCD current from low to high values. By changing the current from high to low values, the transition from high current mode to the low current mode occurs at much lower values of the MHCD current (Fig. 8).

The upper limit in MCS glow discharge current and voltage, respectively, is determined by the onset of the GAT. Then the discharge current rises by several orders of magnitude. Simultaneously the forward voltage drops to a few tens of volts. The $V-I$ characteristics in Figs. 7 and 8 were obtained for discharges in argon at 160 Torr and an electrode gap of 5 mm. The pressure could be increased to 1 atm without reaching the threshold value for the GAT. A side-on photograph of a dc discharge in argon at 1 atm is shown in Fig. 9 between electrodes, which are 2 mm apart. The plasma is bell shaped, with its diameter at the plasma cathode determined by the hole diameter (100 μm). Its diameter increases to 2 mm at the anode. Assuming that the electric field, E, in this discharge is given as the anode voltage divided by the gap distance, and that the current density, j, at midplane is the current divided by the cross section of the discharge at this plane, the electron density at this position can be estimated

$$n_e = j/(E\mu_e e),$$

with the electron mobility, μ_e, being $0.33 \times 10^6\,(\mathrm{cm^2\,Torr/V\,s})$ for argon.[1]

For $p=760$ Torr, $j=0.05\,\mathrm{A/cm^2}$, and $E=1$ kV/cm the estimated electron density at midplane is $7\times10^{11}\,\mathrm{cm^{-3}}$. The cross section of the discharge was obtained from Fig. 9, by considering the radius, where the intensity dropped to half of the maximum intensity, as effective discharge radius.

Plasma Cathode Anode

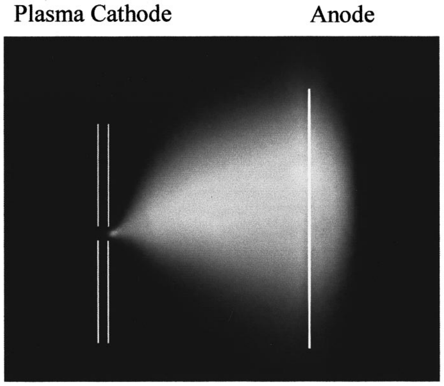

FIG. 9. MCS glow discharge in argon at atmospheric pressure with an anode potential of 213 V. The gap between plasma cathode and anode is 0.2 cm. ($V_{\mathrm{MHCD}}=169$ V, $I_{\mathrm{MHCD}}=470\,\mu$A, $I_{\mathrm{MCS}}=500\,\mu$A.)

V. DISCUSSION

Hollow cathode discharges operate at low current in a Townsend mode, where in an electrode configuration as shown in Fig. 2 (left part) the electric field is dominantly axial. With increasing current they transfer into the hollow cathode mode with high radial electric fields in the cylindrical cathode fall of the discharge. The axial field in the plasma column, which serves as a virtual anode in this case, is rather small. When the hollow cathode discharge operates in this mode external fields generated by a third, positively biased electrode in front of hollow anode can penetrate into the electrode cavity and force the hollow cathode current to flow to the third electrode [Fig. 2 (right part)]. The hollow cathode discharge serves then as electron source for a glow discharge between hollow anode and the third electrode. The hollow cathode system can then be considered as plasma cathode, the third electrode as anode of a MCS glow discharge.

In the plasma cathode sustained glow discharge the cathode fall is eliminated. This is of great importance for the stability of the glow discharge. The cathode fall, a region of high electric field, is generally the cradle of instabilities. Stability is particularly an issue for high-pressure glow discharges, where GATs, emerging from the cathode fall region, are limiting the lifetime of these plasmas to times on the order of microseconds and less. Since in the plasma cathode sustained glow discharge the generation of electrons occurs in the hollow cathode discharge, the glow discharge is stable as long as the hollow cathode discharge is stable, providing that the conditions in the main discharge are such that bulk instabilities are avoided.

It was shown in earlier studies that a stable, high-pressure operation of hollow cathode discharges could be achieved by reducing the cathode hole in the hollow cathode geometry to submillimeter diameter.[6] In argon and xenon these microhollow cathode discharges have been operated, dc, up to and even exceeding atmospheric pressure with cathode hole diameters of 100 μm.[6,9] When used in the three-electrode system as described in this article, we were able to obtain a stable, dc, MCS glow discharge in argon at atmospheric pressure. Electron densities in this glow discharge were estimated to be $7\times10^{11}\,\mathrm{cm^{-3}}$ in the midplane of the main glow discharge.

The volume of the MCS atmospheric pressure discharge is in our experiment still on the order of cubicmillimeter.

However, it should be easy to extend the discharge in axial direction by increasing the applied voltage, as long as the electric field in the discharge is kept below the threshold field for GATs. Extending the discharge in radial direction requires multihole operation, where individual MCS glow discharges are operated in parallel. Experiments in argon at pressures of so far up to 300 Torr have shown that multihole operation of MHCDs is possible. Sixteen discharges have been ignited and sustained in a dc mode by using distributed resistive ballast.[11]

Besides stabilizing a high-pressure glow discharge, the MHCD may also serve as a current valve for the glow discharge. As shown in Fig. 7, the MHCD current determines the glow discharge current at a constant voltage across the main glow discharge gap. The MHCD current on the other hand can easily be controlled by the MHCD voltage. Small variations in this voltage cause large swings in current. The microhollow cathode sustained glow discharge behaves consequently similar as a *vacuum triode:* Small changes in the control voltage cause large changes in the plate (anode) current. This feature of the MCS glow discharge can be used to generate patterns by individually controlling discharges in discharge arrays.

Another feature of the MCS glow discharge, the threshold current required for its onset, opens the possibility to switch a large volume glow discharge on with small voltage swings. By operating the discharge system just below the threshold for onset of the MCS glow discharge (Fig. 7), only a small voltage pulse is required to turn the main discharge on. Once in the on state, it will stay there, even when the voltage pulse is turned off, because of the hysteresis of the main discharge (Fig. 8).

The concept of microhollow cathode discharge sustained high-pressure glow discharges can be applied to any gas. One of the most important gases with applications such as surface treatment and exhaust treatment is air. First results of experiments in air indicate that the stabilizing effect of microhollow cathode discharges on high-pressure glow discharges works also for high-pressure discharges in molecular and electronegative gases.

ACKNOWLEDGMENT

This work was solely funded by the Air Force Office of Scientific Research (AFOSR) in cooperation with the DDR&E Air Plasma Ramparts MURI program.

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