CONF-9509125--

LBL-37218
UC-426

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

Accelerator & Fusion
Research Division

Invited paper presented at the Sixth International Conference on Ion Sources, Whistler, B.C., Canada, September 10–16, 1995, and to be published in the Proceedings

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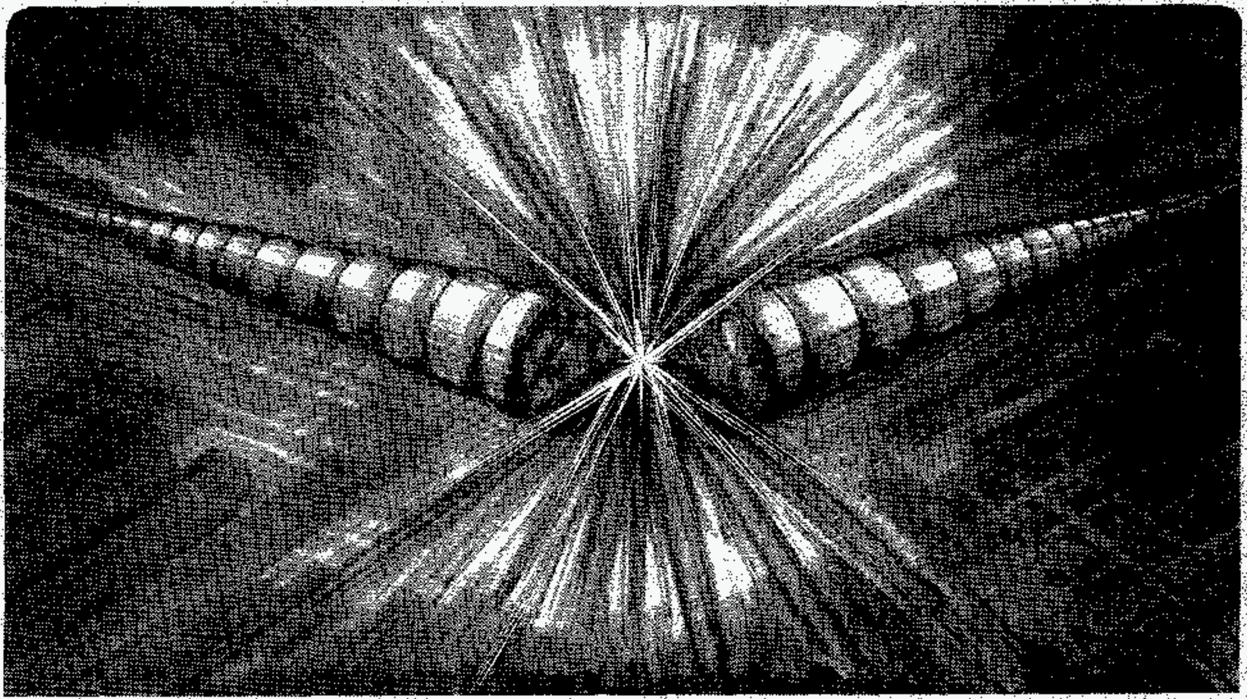
September 1995



Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098

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Version of September 8, 1995

This work was supported by the Office of Energy Research, Office of Basic Energy Sciences, Division of Advanced Energy Projects (BES-AEP), by the Laboratory Technology Transfer Program (ER-LTT), and by the Assistant Secretary for Energy Efficiency and Renewable Energy of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

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Hollow-Anode Plasma Source for Molecular Beam Epitaxy of Gallium Nitride

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Abstract

GaN films have been grown by molecular beam epitaxy (MBE) using a hollow-anode nitrogen plasma source. The source was developed to minimize defect formation as a result of contamination and ion damage. The hollow-anode discharge is a special form of glow discharge with very small anode area. A positive anode voltage drop of 30-40 V and an increased anode sheath thickness leads to ignition of a relatively dense plasma in front of the anode hole. Driven by the pressure gradient, the "anode" plasma forms a bright plasma jet streaming with supersonic velocity towards the substrate. Films of GaN have been grown on (0001) SiC and (0001) Al_2O_3 at a temperature from 600-800°C. The films were investigated by photoluminescence, cathodoluminescence, X-ray diffraction, and X-ray fluorescence. The film with the highest structural quality had a rocking curve with 5 arcmin, the lowest reported value for MBE growth to date.

I. Introduction

Wide band gap semiconductors such as GaN, AlGaN, and InGaN are under intensive investigation for the manufacturing of blue LED's, laser diodes, flat panel color displays, high temperature electronics and other applications. Films of GaN are successfully grown using metalorganic chemical vapor deposition (MOCVD). Molecular beam epitaxy (MBE) represents an alternative approach which might be advantageous to MOCVD due to lower growth temperature and because post-processing is not necessary to activate dopants. The growth by MBE, however, occurs under thermodynamically unstable conditions, and it is crucial to drive the forward synthesis reaction between gallium and nitrogen by providing a sufficient flux of activated nitrogen species with low kinetic energy [1, 2]. An ideal source of activated nitrogen species (N, N^*, N_2^*, N_2^+, and N_2^{+*}) would have a number of features including UHV compatibility, high flux of activated species, extremely low contamination level (in the ppb range), low kinetic energy of all species, stable operation, and simple maintenance.

Activated species are usually provided by ECR plasma sources [3-6], or by microwave or RF plasma sources [7, 8]. In both cases, the energy distributions have a significant contribution of undesired high energy species. Better energy control is obtained by using a Kaufman ion source [1, 2, 9] operating at its low energy limit. Often, substrate bias is used to have additional control on the energy of ionized species [1-3, 8].

This paper is the first report about MBE growth of GaN films using a hollow-anode plasma source. We focus here on the features of the source, and the properties of the GaN films will be described in more detail elsewhere.

II. Hollow-anode plasma source

The hollow anode plasma source employs a modified glow discharge with a very small hollow anode. The first basic ideas for a hollow-anode ion source have been published by Miljevic [10-12] in the mid 1980s. Miljevic used originally a concave cathode but it was later shown that this feature is not essential [13]. The working principle of the source was recently investigated in [14], and we give here a brief summary of this work.

The basic arrangement of a hollow-anode discharge is shown in Fig. 1. Nitrogen is fed through an opening in the cathode. A glow discharge is established between cathode and anode which are separated by an insulator (a ceramic cylinder in our case). The nitrogen plasma exits the discharge chamber through a hole in the anode, driven by the pressure gradient. The anode area is kept small by introducing an insulating barrier in front of the anode, thus making the small outlet opening the only active anode area. A glow discharge can be sustained without the anode barrier, but a bright plasma flow is only observed if the barrier is installed (see below).

Typical dimensions and parameters of the source are: anode-cathode distance 2-10 cm, anode hole opening 0.5 - 2 mm, gas flow rate 1-100 scc/min, pressure inside the source 10-100 Pa ($0.1-1\,\mathrm{Torr}$), pressure outside the source $10^{-3}-10\,\mathrm{Pa}$ ($10^{-5}-10^{-1}\,\mathrm{Torr}$), depending on anode opening and pumping speed, applied voltage (including voltage drop at the current-limiting resistor of 2-5 kΩ) 600 - 1000 V, self-sustained cathode-anode voltage (during operation) 450 - 800 V, discharge current 20-200 mA, ion "beam" current 0.1 - 10 μA.

The plasma properties of the hollow-anode plasma source have been investigated in Ref. [14] by using a movable Langmuir probe which was located inside the source. The plasma parameters (density, electron temperature, floating potential and plasma potential) were determined from probe characteristics. An additional flat Langmuir probe was used to measure the properties of the plasma leaving the source.

If the source was used without the insulating barrier (large anode area), the nitrogen plasma flow was very weak and the anode voltage drop was about 15 V. The anode sheath thickness was smaller than 2 mm. In contrast, if the anode barrier was used, a very bright plasma region at the anode hole was observed, and the outflowing nitrogen was clearly ionized and excited (Fig. 2). Several of our sources had two stages in series; the plasma jet of the first stage helped to maintain the plasma production in the second stage at relatively low burning voltage. The source in Fig. 2 is such a dual-stage source, and the insulators were made of glass for better visual observation. The plasma streams to the right and is clearly visible not only inside but also outside the source.

Figure 3 shows the plasma potential and electron density as a function of distance to the hole center in the anode plane for two different flow rates. It was found that (i) the anode potential drop is significantly greater than without the insulating anode barrier, (ii), the anode sheath thickness is increased, and (iii), the increase in density is faster than exponential when approaching the anode hole. The electron temperature was in the range between 1 and 3 eV, with the lower values in the high-density region. The gas pressure inside the plasma source, the discharge current, anode-cathode voltage and all plasma parameters depend on the flow rate. Keeping the supply voltage constant, a maximum in the discharge current (180 mA), the anode drop (about 40 V), plasma brightness, and plasma output was observed at a flow rate of about $50\ \mathrm{scc/min}$. An ion density in the range $n_i = 2-8\times10^8\ \mathrm{cm}^{-3}$ was found at 8 cm distance from the plasma source. This corresponds to a particle flow density of $1-4\times10^{11}\ \mathrm{ions/cm^2\ s}$. Note that the particle flow density can be enhanced by increasing the plasma source power and operating at the optimum flow rate (the "optimum" flow rate is obtained when the discharge current shows a maximum).

III. Growth of GaN using a hollow-anode plasma source

A UHV-compatible dual-stage hollow-anode plasma source was specifically built for the growth of GaN. The source is relatively simple in design and has water-cooling for stable operation. To date, the source has operated in the MBE chamber for more than half year. Only high-purity materials have been used: aluminum 1100 F series (purity better than 99.0%) for the electrodes and AlN ceramics for the insulating parts. In this way the source is exclusively built with compatible materials which do not degrade the optical and electrical performance of GaN films.

The anode of the source was grounded, and thus the plasma leaving the source has a plasma potential very close to ground. The kinetic energy of the species results from gasdynamic acceleration in the anode hole. The anode opening acts like a nozzle, and supersonic velocities of Mach number 1.2 have been observed [14]. The associated kinetic energy is less than 1 eV, and additional ion acceleration or deceleration can be achieved by negative or positive substrate biasing, respectively. A more detailed analyses of the kinetic energy of the species in the plasma is in progress.

The GaN films in this experiment were grown in two MBE chambers equipped with Ga Knudsen cells, heated substrate holders and cryoshields. Both chambers were cryopumped (base pressures in the 10^{-10} Torr range). The supply gas was 99.9999% pure nitrogen flowed through a Nanochem purifier. 1 cm x 1 cm (0001) SiC and (0001) Al_2O_3 substrates were used. Indium was deposited on the back of each substrate as a thermal contact. Growth was initiated with a 10 nm buffer deposited at 500°C. The substrate was then ramped up to the growth temperature of 600-800°C.

Photoluminescence (PL) was performed at 4 K using an Omnichrome 30 mW HeCd laser ($\lambda = 325$ nm). To measure the spatial homogeneity of the grown films, cathodoluminescence (CL) was performed using a JEOL 35 CF electron microscope with a cooled liquid-nitrogen stage. The cathodoluminescence spectrum at 95 K is dominated by near band-edge donor-bound exciton emission at 360 nm emission with a FWHM of 20 meV. Mid-gap emission at 550 nm which is commonly observed in GaN is found to be at least 1000 times smaller than the near-bandedge emission. The lateral homogeneity of the optical properties for films grown on sapphire were significantly improved over films grown with the Kaufman ion source. With the Kaufman source, a positive bias was applied to the substrates in order to reduce the kinetic energy of the ions and reduce ion damage. The substrates were held in place with molybdenum clips. Since sapphire is electrically insulating, there was a large voltage drop at the edges of the film, near the clips. Thus, the advantageous effects of the positive bias only occured at the edges. This resulted in optically dead material in the middle and strong near band-edge donor-bound exciton emission at the edges. In contrast, the hollow-anode source produced homogeneously strong luminescence due to the inherently low kinetic energy of the plasma species.

Structural characterization was performed using a Siemens D5000 X-ray diffractometer with Cu-K α radiation, four-circle translational capability and four-bounce Ge monochromator. Normal-coupled ($\Theta - 2\vartheta$) and rocking-curve X-ray diffraction data indicate that the films are single phase with the hexagonal structure and the c-axis parallel to the growth direction. The film with the highest structural quality was grown on sapphire directly without a buffer layer. This film was found to exhibit a rocking curve FWHM of 5 arcmin. This is the lowest reported value for MBE growth to date.

Trace impurity analysis was performed using X-ray fluorescence at the microbeamline of the Advanced Light Source at Lawrence Berkeley National Laboratory. The presence of Al below several percent is not considered deleterious to film quality since Al is isoelectronic to Ga and therefore does not result into an increase in deep level defects within the films. Instead, a practically insignificant shift in the bandgap energy occurs, without degradation to device performance. The contamination of the films grown with the hollow-anode source was significantly improved over earlier MBE work.

IV. Summary and outlook

A UHV-compatible hollow-anode plasma source has been specifically developed for MBE of GaN thin films. The source is made only of Al and AlN, i.e., materials which do not result in degradation of GaN film quality. The kinetic energy of the species in the downstream plasma flow is very low since plasma acceleration is purely gasdynamic. Use of this source leads to a reduction of defects in GaN films which is attributed to both low contamination levels and the low kinetic energy of activated nitrogen. Most remarkable is a rocking curve with 5 arcmin FWHM, the smallest value obtained by MBE of GaN.

The development of a more compact version of the source is under way, improving it in terms of output and simplicity. It was shown that the source can operate with other gases, too, and a number of other applications are anticipated.

Acknowledgments

The authors would like to thank L. Schloss, J. Chan, X. Liu and E. Weber for assistance with characterization of GaN. This work was supported by the Office of Energy Research, Office of Basic Energy Sciences, Division of Advanced Energy Projects (BES-AEP), by the Laboratory Technology Transfer Program (ER-LTT), and by the Assistant Secretary for Energy Efficiency and Renewable Energy of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

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Figure Captions

Fig. 1 Basic arrangement of a hollow-anode discharge.

Fig. 2 Test version of the hollow-anode plasma source in operation (dual-stage version, not UHV compatible, with optional magnetic field); the plasma streams to the right.

Fig. 3 Plasma potential and electron density inside the source as a function of distance to the center of the hole in the anode plane.

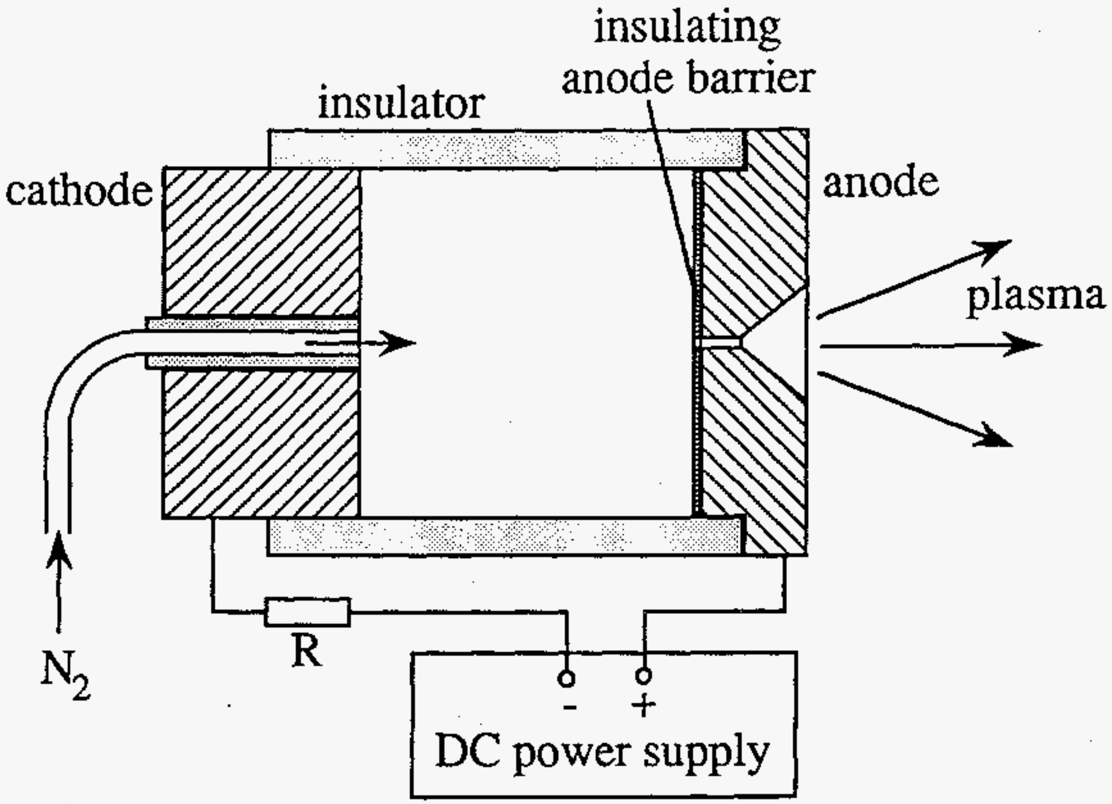

insulating
anode barrier
insulator
cathode
anode
plasma
N_2
R
-
+
DC power supply

Figure 1

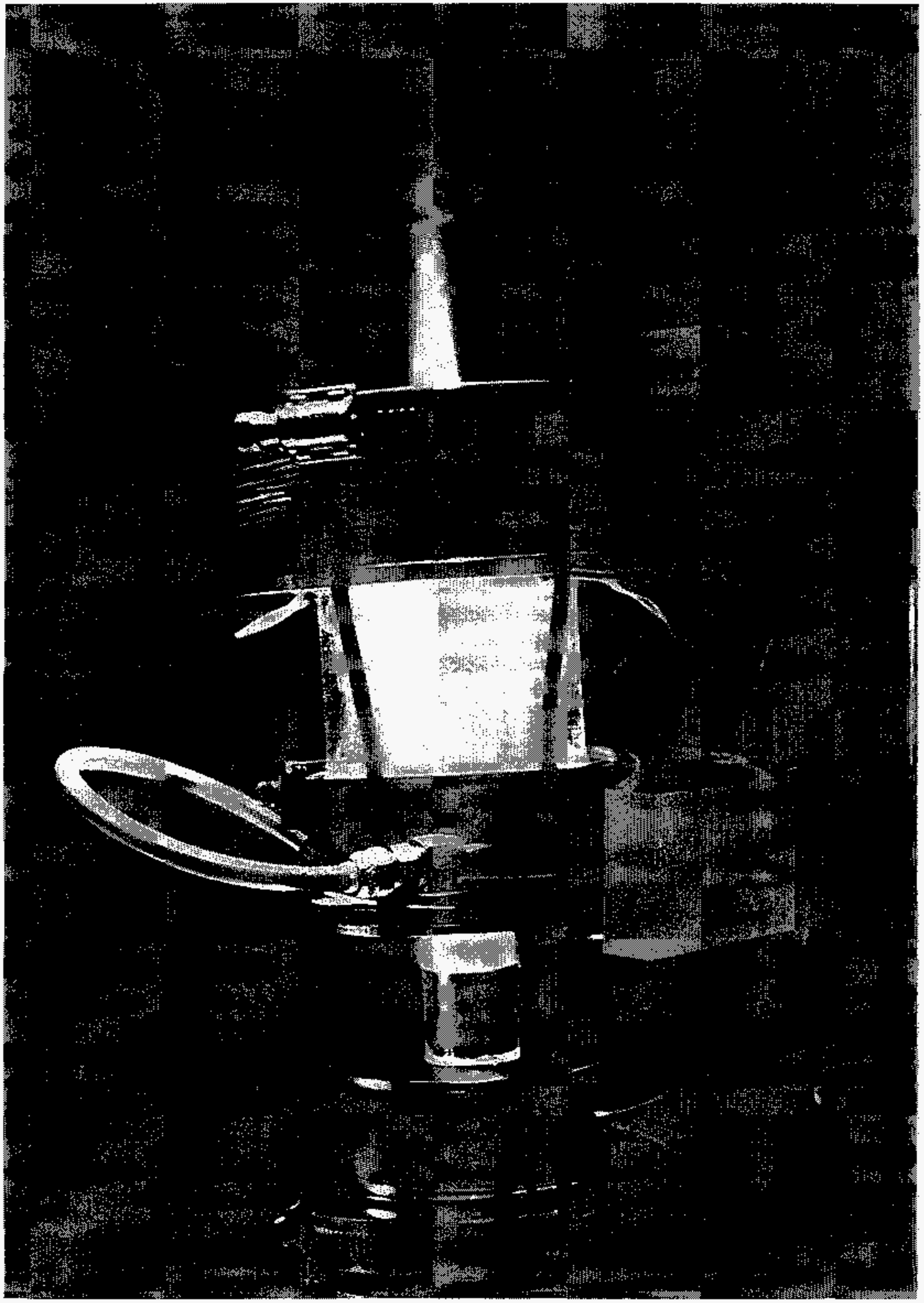
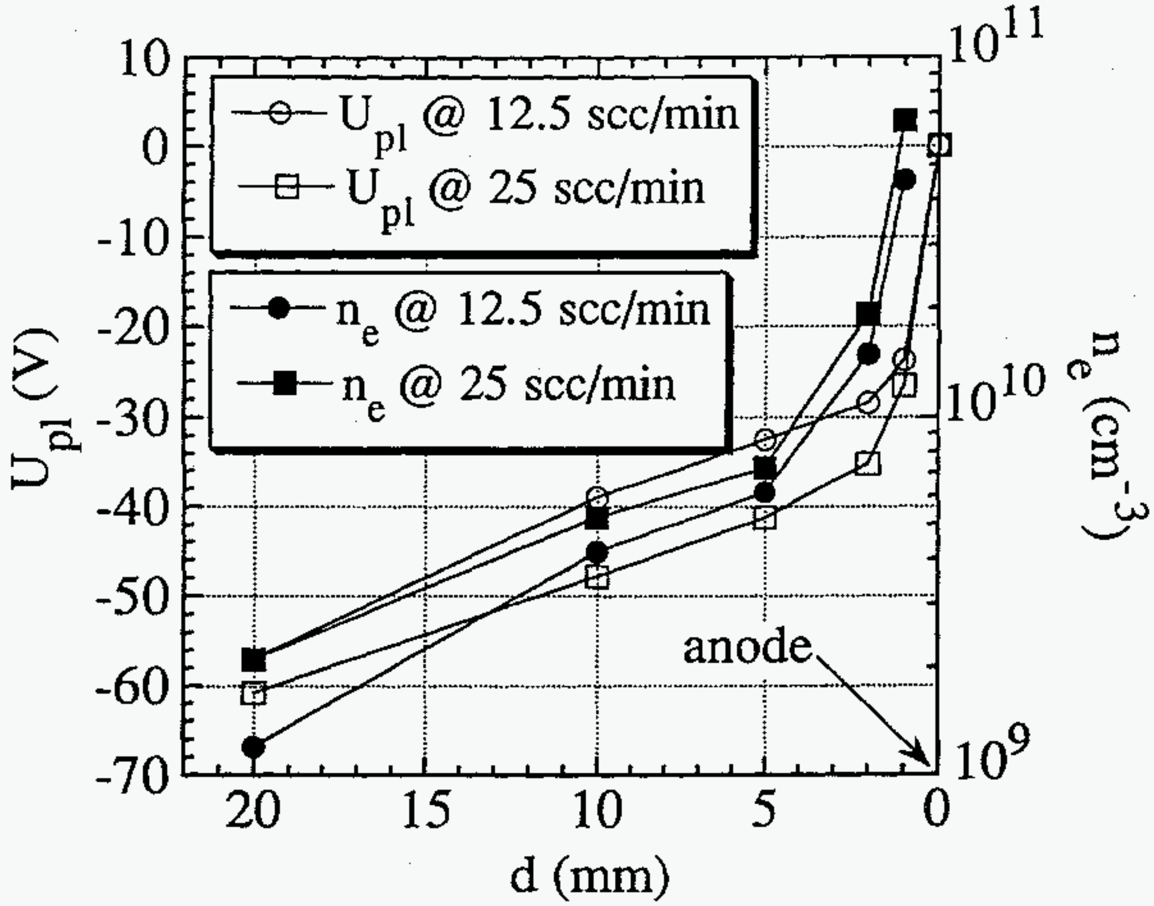


Figure 2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U_pl @ 25 scc/min
n_e @ 12.5 scc/min
n_e @ 25 scc/min
U_pl (V)
n_e (cm^-3)
d (mm)
anode
10
0
-10
-20
-30
-40
-50
-60
-70
20
15
10
5
0
10^11
10^10
10^9

Figure 3