I. INTRODUCTION

The positive column of a glow discharge in a molecular gas is an useful source of highly non equilibrium populations of excited molecules and atoms. Steady-state discharges have been the subject of numerous investigations [1] [3] but theoretical and experimental studies devoted to flowing gas discharges are relatively rare and nearly all of them are restricted to rare gases [4] [5]. The objective of the present work was to develop a model for predicting the properties of the positive column in a glow discharge in flowing nitrogen. The model is similar to that previously used to describe a glow discharge in flowing hydrogen [6]. However, in contrast to hydrogen discharges where direct ionization predominates, nitrogen ionization in glow discharges arises mainly from multistep ionization processes. Among them, associative ionization produced by collisions between metastable molecules, one in the a’1Σu+ state and the other in the same state or in the A3Σg+ state [7], appears to be dominant at low and moderate pressures. This ionization mechanism was used in the present model and comparisons were made with past experimental data and with our own observations in order to test its validity. Good agreement between theoretical and experimental results were observed at low and moderate pressures for nitrogen flow with low Reynolds numbers.

II. DISCHARGE MODEL

The positive column of a cylindrical glow discharge in flowing nitrogen is considered. The plasma is assumed to be weakly ionized and collision dominated. Charge neutrality is assumed so that \( n_e = n_i \) where \( n_e \) and \( n_i \) denote electron and positive ion density, respectively. Ambipolar diffusion as well as equilibrium of the positive ions with the gas is also assumed. As the axial flow velocity \( u \) is always much smaller than the electron drift velocity \( \mu_n E_z \), the equation for the conservation of electrons can be written:
1) \[ \frac{1}{r} \frac{\partial}{\partial x} \left( r \left( n_e v_{\mu_1 U_e} \frac{\partial n_e}{\partial x} \right) \right) - \frac{\partial}{\partial x} \left( n_e \mu_e E_x \right) = S_1 \]

where \( v \) is the radial gas flow velocity, \( n_e \) the electron density, \( n_i \) the ion density, \( U_e \) the electron characteristic energy, \( \mu_1 \) the ion mobility, \( \mu_e \) the electron mobility and \( E_x \) the axial electric field, respectively. The source term \( S_1 \) is written in the form:

2) \[ S_1 = k_i N_2(X)n_e + k_{ii}a'A + k_{ii}a'^2 - \alpha_r n_e^2 \]

where \( k_i N_2(X) \) represents the rate at which electrons are produced by direct ionization, \( k_{ii} a'A \) and \( k_{ii} a'^2 \) the rate at which they are produced by two-step ionization according to the mechanism previously discussed (7) and \( \alpha_r n_e^2 \) the rate at which they are lost by recombination. In these terms, \( k_i \) is the electron ionization rate, \( k_{ii} \) and \( k_{ii}' \) the association ionization rates, \( \alpha_r \) the electron recombination rate, \( a', A \) and \( N_2(X) \) are the number density of molecules in the \( a'^1 \Sigma^+_u, A^3 \Sigma^+_u \) and \( X^1 \Sigma^+_g \) (fundamental) electronic states, respectively.

In Eq. 1, the axial electric field \( E(x) = E_x \) takes a value such that the current conservation is satisfied.

3) \[ I(x) = 2\pi n_e \int_0^R \mu_e(x) E(x) rdr = I_0 \text{ constant} \]

It should be noted that at first sight, the electric field should be the vacuum field since quasi-neutrality of the plasma has been assumed. Therefore the electric field should be constant in most part of the discharge beyond the electrode regions. However, it was found that the electric field decreases along the discharge axis. This is due to a very small charge separation (\( n_e/n_i - 1 = 10^{-6} \)) which skews the vacuum field in order to preserve the current continuity according to Eq. 3. Therefore for a constant current \( I_0 \), the unknown \( n_e \) and \( E \) can be determined by solving simultaneously Eqs. 1 and 3.

The coefficients \( k_i, k_{ii}, k_{ii}', \mu_e, U_e \) and \( \alpha_r \) used in Eqs. 1-3 are functions of \( E/N \) and the next step of the calculation was to determine \( N \) from the conservation equations for mass, momentum, energy and species of the gas flow. These equations were written in a form similar to that used for modeling chemical lasers [8] and they have been already presented in a simplified form [6]. Vibration-vibration (V-V) exchange processes and vibrational deactivation by electrons were considered for vibrationally excited nitrogen molecules.

For the electronically excited molecules and the \( N \) atoms, the following processes were considered:

\[ N_2(A) + N_2(A) = N_2(B) + N_2(X) ; K_{AB} = 1.1 \times 10^{-9} \text{ cm}^{-3} / \text{s} \]
\[ \begin{align*}
N_2(A) + N_2(A) & \xrightarrow{K_{AC}} N_2(C) + N_2(X) ; K_{AC} = 2.5 \times 10^{-10} \text{ cm}^3/\text{s} \\
N_2(A) + N_2(X) & \xrightarrow{K_{AX}} 2N_2(X) ; K_{AX} = 3 \times 10^{-18} \text{ cm}^3/\text{s} \\
N_2(B) + N_2(X) & \xrightarrow{K_{BX}} N_2(A) + N_2(X) ; K_{BX} = 2 \times 10^{-12} \text{ cm}^3/\text{s} \\
N_2(A) + N_2(a') & \xrightarrow{k_{11}} N_2^+ + N_2 + e ; k_{11} = 5 \times 10^{-11} \text{ cm}^3/\text{s} \\
N_2(a') + N_2(a') & \xrightarrow{k_{11}'} N_2^+ + N_2 + e ; k_{11}' = 2 \times 10^{-10} \text{ cm}^3/\text{s} \\
N_2(a') + N_2(X) & \xrightarrow{K_{AX}} 2N_2(X) ; K_{AX} \approx 6 \times 10^{-14} \text{ cm}^3/\text{s} \\
N + N + N_2(X) & \xrightarrow{K_R} N_2(B) + N_2(X) ; K_R = 2.4 \times 10^{-33} \text{ cm}^6/\text{s}
\end{align*} \]

III. COMPARISON WITH EXPERIMENTAL RESULTS AND DISCUSSION

POLAK et al [15] carried out a complete set of measurements in a 3.2 cm diameter, 30 cm long discharge at pressures of 0.65 - 6.3 Torr, currents of 5 - 75 mA and gas flow velocities of 0.5 - 1.0 m/s. They measured the longitudinal electric field, the gas temperature on the axis and at the walls of the discharge tube, the electron density and the populations of the \( N_2(A^3L_u) \), \( N_2(B^3\pi_u) \) and \( N_2(C^3\pi_u) \) electronic states. They also determined the volume ionization rate for various discharge conditions and they suggested an ionization mechanism to explain their experimental results. Extensive comparison between our model predictions and their observations was made at pressures of 2.0 and 3.9 Torr and gas flow velocity of 1 m/s (Reynolds number \( \approx 20 \)).

Of particular interest in the results of the calculations are the variation of the vibrationally and electronically excited nitrogen molecules along the discharge axis. It was found that the populations in the higher vibrational levels rise slowly in contrast to those in the electronic states which rise rapidly near the origin. Therefore the associative ionization mechanism suggested by POLAK and involving vibrationally excited molecules in levels \( v > 13 \) should be ruled out since it cannot explain the high rate of volume ionization observed at low \( E/N \) values.

Predicted and measured values of the electric field were found in good agreement. This supports the associative ionization mechanism involving metastable molecules in the \( a^3\Pi_u \) and \( A^3\Pi_u \) electronic states. Comparison with other experimental data show good agreement between predicted and measured values for discharge at low and moderate pressures.
REFERENCES

[1] G. ECKER and O. ZOLLER
Phys. Fluids, 7, 2001 (1964)

J. Appl. Phys., 44, 4920 (1973)

Phys. Fluids, 19, 819 (1975)


AIAA Journal, 13, 647 (1975)

J. Physique 42, 1525 (1981)

(1985)

"Analysis of Chemical lasers", Report RK-CR-74, Lockheed Missiles


High Temperature, 15, 13 (1977)