"LOCAL" VIBRO-ROTATIONAL ANALYSIS OF NON-EQUILIBRIUM NITROGEN PLASMA AT MODERATE PRESSURES

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ABSTRACT

A "local" vibro-rotational analysis of the excited species produced in flowing nitrogen r.f. discharge has been carried out by measuring radial and axial emission intensities of some vibrational sequences and selected rotational lines of the (0,2) band of the SPS of N_2 ($C^3\pi_u$ - $B^3\pi_g$).

General maps of emission intensities, and of vibrational and rotational temperature distributions have been derived.

1. INTRODUCTION

The spectroscopic diagnostics of electrical discharges of the type utilized in our laboratory for chemical application (1-4) have been found to be a powerfull tool for revealing the existence of non-equilibrium conditions between the various degrees of freedom (4-5) and for establishing the relationship between the discharge parameters and the kinetic mechanisms of excitation and deexcitation of the chemical species present in the plasma. The present analysis has been carried out with the aim of a detailed axial and radial characterization of the chemical reactor.

2. EXPERIMENTAL

Details of the experimental apparatus can be found in ref. (4-6). The optical system is essentially made of a plane mirror, which can rotate its axis and of a spherical lens which focusses the central longitudinal plane of the plasma column on the entrance slit of a 1 m Jarrel-Ash monochromator. The axial translation of the plasma column and the rotation of the plane mirror allow the radial and axial analysis of the cylindrical reactor. Pure nitrogen, at a fixed flow of 400 $cm^3(STP)min^{-1}$, has been utilized, in the pressure range 5-35 torr, with approximately constant power input of 3 watts cm^{-3}. Three axial positions of the plasma column have been analyzed, at distance of 5, 55 and 105 mm, respectively, from the lower edge of the upper electrode, with a fixed electrode separation of 110 mm.

Side on measurements of the relative vibrational band and rotational line intensities I(x) have been taken at fixed lateral position x across the plasma section. The radial intensity profile, $\varepsilon(r)$, of each vibrational band head or rotational line of N_2(SPS) have been obtained with the Abel's integral equations, which have been solved using the numerical approximation of Bockasten (6-7). Radial vibrational (T_V) and rotational (T_R) temperature have been obtained with the "Boltzmann plot" technique. General maps of the normalized emission intensities and vibrational temperature distributions within the reactor derived from this analysis are shown in fig. 1, a and b, respectively.

3. RESULTS AND DISCUSSION

The main results of the analysis can be summarized as follows. - 1) The temperature gradients between the core and the periphery of the discharge are less steep than the relative gradients of emission intensity; - 2) The inversion process enhances the temperature gradients present in the profile derived from the lateral intensities; - 3) The maxima in the emission intensity are not related to the maxima in the temperature profiles; - 4) The effect of pressure is different for the three axial positions, but in general T_V tend to decrease and T_R to increase with increasing the pressure; - 5) The values of T_V in the plasma core are almost independent on the axial position, except in the vicinity of the upper electrode, where the plasma column is strongly affected by the funnel shaped electric field; - 6) The maximum values of T_V are slightly out of the discharge axis, and appreciable increases are observed at the border of the discharge; - 7) The values of T_V range from 4500-5000 K, in the core, to 2500-3000 K, at the border, while T_R can vary from ~ 4500 K to 1000 K, from the core to the periphery.

The radial intensity profiles, are not related in any way to the temperature profiles. Variations in the band intensity profile of the order of 10^3 correspond to variations of T_V only of less than a factor two. Moreover, while the emission intensity decreases drastically at the border of the discharge, T_V tends to increase, especially in the central part of the reactor. The results of this analysis have been interpreted on the basis of the following arguments.

The vibrational energy distribution for the ground electronic state $X^1\Sigma_g^+$ is determined by e-V and V-V energy exchange processes (3). For the e-V processes, the rate constant is a function of the cross sections, $\sigma(u)$, which have significant values only for electron energy less than 2 eV, and of the electron energy distribution function f(u), a unique function of the electric field. Slight variations in electron density from the core to the periphery of the discharge can lead to small gradients in the values of T_V, while gradients in the electric field would effect only to a minor extent than part of f(u) which is relevant to e-V processe (8-9). The behaviour of T_V in the $C^3\Pi_u$ state should reflect the corresponding distribution of the ground state (10).

For the emissivity one should examine the variation of the number density of the $C^3\Pi_u$ state, from which the SPS originates. If the principal mechanism of excitation is the direct electron impact from the ground state, the cross section for the process has a threshold at 11 eV. For this process, therefore, is the tail of f(u) to determine the value of the rate coefficient and then the number density of $C^3\Pi_u$ state. Since the tail of f(u) is strongly affected by small variation of the electric field, a remarkable decrease of the $C^3\Pi_u$ emitters can results in going from the core to the periphery of the discharge column.

The observed increase of T_V at the border of the plasma, where the emission intensity is extremely low, should be attributed to the presence of additional mechanisms of $C^3\Pi_u$ population, very likely involving energy exchange between electronic excited state (11), which can lead to molecules in the $C^3\Pi_u$ state with a vibrational energy distribution slightly higher than that obtained by electron impact process.

The pressure dependence of T_V and T_R has been interpreted on the basis of vibrational energy relaxation from high v-states in favour of rotational energy by means of pressure dependent very fast processes (12).

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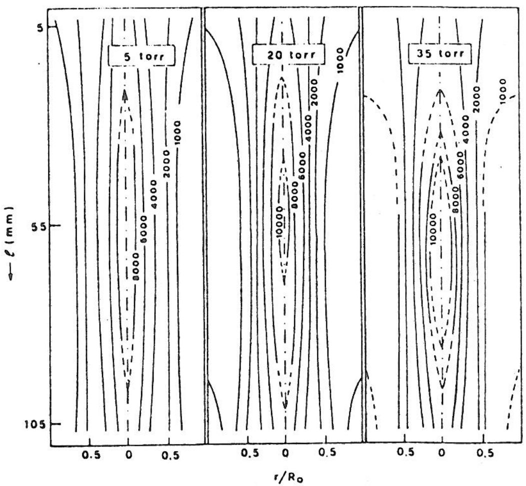
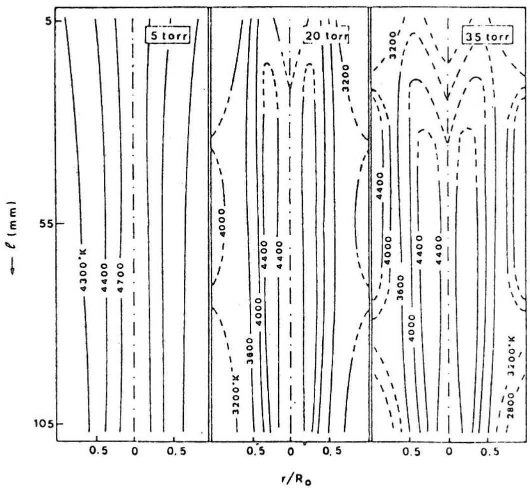
Fig. 1



a) Radial intensity maps (in arbitrary units), at different pressures.



b) General maps of the radial T_V of $N_2(C^3\Pi_u$ state) within the reactor at different pressures.