Study of N_2/He DC glow discharge

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Abstract. Optical emission spectroscopy and mass spectrometry measurement were used to study a gas mixture glow discharge of He and N_2 at total pressure of 2.0 Torr, a power of 10 W and a flow of 16.5 l/min. The emission bands were measured in the wavelength range of 200 to 1100 nm. The principal species observed were N_2^+ $(B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+)$, N_2 $(C^3\Pi_u \rightarrow B^3\Pi_g)$, and He, which agree with mass spectrometry measurement.

1. Introduction

Recently, various kinds of plasma have been produced to obtain excited states, active atoms and molecules. The production of active species is very important for a multitude of applications, such as biological sterilization and heating, plasma-assisted ignition and combustion, surface treatment and destruction of harmful substances and pollutants [1-9]. It is well known that an admixture of a molecular gas to a noble gas can substantially change the electrical and spectral characteristics of a glow discharge. The degree and way of these changes depend on the discharge conditions: the sort of noble gas, sort and percentage of the admixture, gas pressure, etc. Here, we restrict ourselves to the analysis of the present state of the art in studying DC glow discharge of a N_2/He mixture.

This paper is intended to acquire knowledge of emission spectroscopy and mass spectrometry in a gas mixture glow discharge of N_2 and Ar, at total pressure of 2.0 Torr. The OES and mass spectrometry dependence on the concentration of the mixture are presented and discussed.

2. Experimental details

The schematic diagram of the experimental setup is identical to that described in previous work [10, 11]. The discharge cell consists of two parallel electrodes enclosed in stainless steel vacuum chamber. The two electrodes were made of stainless steel disc, with 30 mm in diameter. The electrodes are positioned at the center of the reaction chamber with 10 mm gap spacing. The plasma chamber had a volume of 1.16 x 10^4 cm^3 and it was pumped down by a vacuum system (Turbomolecular pump Varian D5302) to a base pressure of 10^{-6}Torr. A continuous dynamic flow of He -N_2 gas mixture (ultra-pure gases, Praxair 99.99%) was let in the system through needle valves at the desired pressures. A DC glow discharge was produced between the two electrodes. While keeping the total pressure of 2.0 Torr, the concentration of He gas in the mixture was done by changing the He partial pressure. To keep a current constant of 10 mA (measured by a digital Fluke multimeter model 8846A), independent of the gas mixture, a ballast resistance (R=5 KΩ) was used, it was done by changing the power supply voltage (Spellman SA4) between 260 V (for 100 % of He) and 250 V (for 100 % of N_2). The discharge voltage in gas mixture is lower than that in pure Helium. A lateral flange was a quartz window, used to monitor the active species generated in the glow discharge by plasma emission spectroscopy; the

optical spectroscopy measurements (OES) were carried out by using a high-resolution Ocean Optics Inc. Spectrometer HR2000CG-UV-NIR and a UV2/OFLV-5 detector, the fiber optic was placed in the aperture entrance. The grating has a spectral response in the range of 200 to 1100 nm.

A quadrupole mass spectrometer (QMS) is connected via cutoff and needle valves to the recipient for process gas analysis by in situ mass spectrometry (MS). The process gas composition and its changes during N_2/He plasma are measured using Faraday cup detection mode at a pressure of 5.0×10^{-5} Torr inside the analyzer of the mass spectrometer. Ion currents (I_m) of several mass (m) are recorded simultaneously versus He % in the mixture of the plasma.

In preparation for every MS measurement, the mass spectrometer is pumped down. A final pressure of 1.5×10^{-6} Torr is reached afterwards.

3. Results and discussion

OES measurement of glow discharge plasma at a pressure of 2.0 Torr is displayed in Fig. 1. This allowed analysis of the most luminous area, which corresponds to the negative glow near the cathode dark space. Only the most intense spectral lines and bands of the plasma within the 200-1100 range are quoted [12]. The principal species observed were: at 337.13 and 357.69 nm for N_2 ($C^3\Pi_u - B^3\Pi_g$); at 391.44 and 427.81 nm for $N_2^+ (B^2\Sigma_u^+ - X^2\Sigma_g^+)$; at 501.57, 587.56 and 667.82 nm for He.
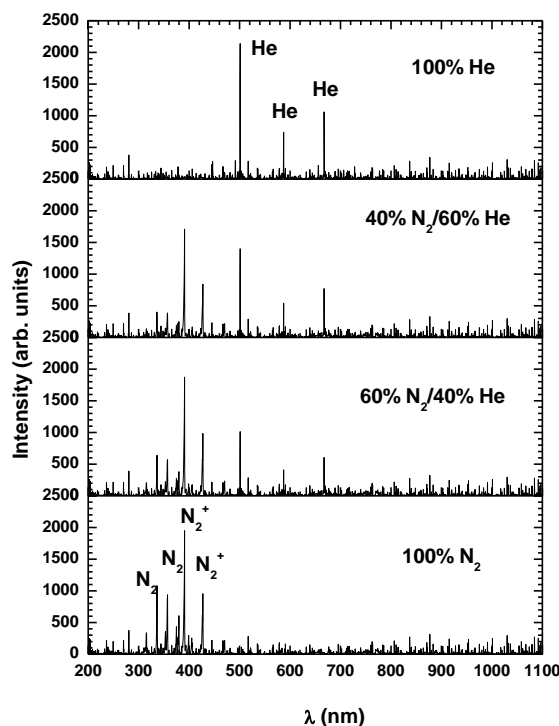

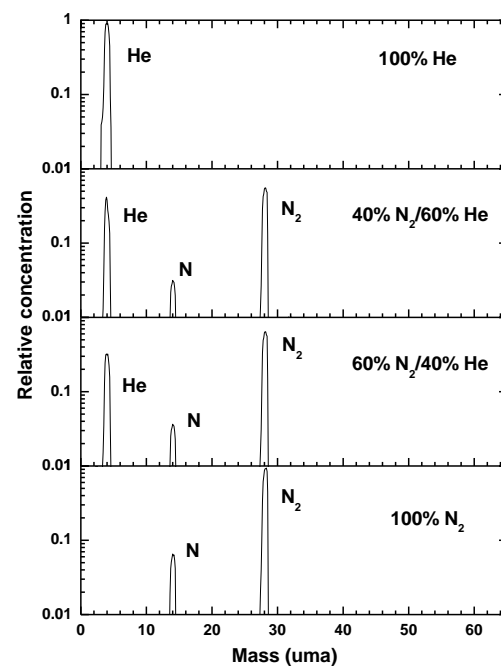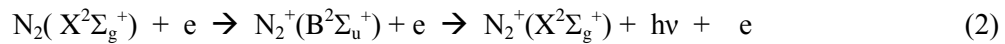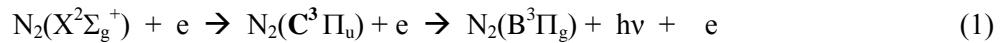Figure 1. Emission spectra of N_2-He plasma

Figure 2. Relative concentration of neutral species.

Figure 2 shows the relative concentrations of three masses as functions of He % in the mixture of the N_2-He plasma at 2.00 Torr. The experimental results display three species of stables neutral, which can be associated to the N_2 molecule and, N and He atoms.

From figures 1 and 2, it can be seen that both shown the same behaviour of the species observed as a function of the He percentage, with the only difference that in the MS measurement, it observes a relatively low intensity corresponding to N atoms, which decreases as the He percentage increases.

Under our experimental conditions the important processes of inelastic electron-atom, electron-molecule and subsequent emission transitions from excited states are

$$N_2(X^2\Sigma_g^+) + e \rightarrow N_2(\mathbf{C^3}\Pi_u) + e \rightarrow N_2(B^3\Pi_g) + h\nu + e \tag{1}$$

$$N_2(X^2\Sigma_g^+) + e \rightarrow N_2^+(B^2\Sigma_u^+) + e \rightarrow N_2^+(X^2\Sigma_g^+) + h\nu + e \tag{2}$$

$$He(1^1S) + e \rightarrow He(3^1P_1) + e \tag{3}$$

$$He(1^1S) + e \rightarrow He(3^3D_{1,2,3}) + e \tag{4}$$

$$He(1^1S) + e \rightarrow He(3^1D_2) + e \tag{5}$$

The excitation energy of the emitting state $N_2(\mathbf{C^3}\Pi_u)$ at 337.13 nm is 11.03 eV and the emission cross sections for electron impact excitation of the nitrogen molecule are in the order of 10^{-22} to 10^{-23} m^2, while the excitation threshold for the $N_2^+(B^2\Sigma_u^+)$ band at 391.44 nm is about 18.75 eV above the ground state of the neutral molecule, and the corresponding cross section for optical emission caused by electron impact approaches 2 x 10^{-22} m^2 for electron energies around 50 eV [13]. Because of its high intensity as can be seen in Figure 1, this band was preferentially used in combination with band from the neutral second positive system for an evaluation based on the corona model [14].

In N_2/He discharges N_2^+ ions are mainly produced by Penning ionization of $N_2(X^2\Sigma_g^+)$ by metastable He atoms. According to data published by Hotop et. al. [15], about 40% of these ions are formed in the state $N_2^+(B^2\Sigma_u^+)$. Production of $N_2(X^2\Sigma_g^+)$ by charge transfer between helium ions and N_2 molecules is not effective under our experimental conditions. The rate constant of this process is high 1.2 x 10^{-9} cm^3/molecule s^{-1} [16] as compared to $\approx 10^{-10}$ cm^3/molecule s^{-1} for Penning ionization process. But the production rate of the He ions is two orders of magnitude smaller than the production of metastable helium atoms. Moreover, it was shown by Collins [17] that N_2^+ is produced in He/N_2 mixtures mostly by Penning ionization process, while charge transfer gives nitrogen atoms in a dissociative or predissociative state, which it can be seen in the MS measurements (see Figure 2).

4. Conclusions

The experimental investigation of cold plasma in which N_2 is predominant can be of great interest for identification of species and processes of likely relevance in multitude of applications, such as biological sterilization and heating, plasma-assisted ignition and combustion, surface treatments and destruction of harmful substances and pollutants.

In this work, we studied by optical emission spectroscopy and mass spectrometry measurements of the plasmas formed in low-pressure (2.0 Torr) glow discharge of N_2 with admixtures of He.
The principal species observed were N_2^+ ($B^2\Sigma_u^+\rightarrow X^2\Sigma_g^+$), N_2 ($C^3\Pi_u\rightarrow B^3\Pi_g$), and He, which agree with MS measurement.
It was shown that N_2^+ is produced in He/N_2 mixtures mostly by Penning ionization process, while charge transfer gives nitrogen ions in a dissociative or predissociative state, which has been corroborated by MS measurements.

Acknowledgments

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