

Decaying of Nitrogen Second Positive System by Addition of H₂ Gas in Air DB Discharge

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ABSTRACT

Effects of addition of H₂ gas in air DB discharge on its optical and electrical characteristics have been studied. Optical emission spectroscopy is used to investigate the effect of hydrogen admixing on the emission intensity of the nitrogen second positive systems (300 - 420 nm) and the relative population density of N₂(C³Π_u, ν') states. An obvious decaying of the emission intensity of the nitrogen second positive bands with the introduction of H₂ has been observed. It has been concluded that quenching of the nitrogen excited state N₂(C³Π_u, ν') is the responsible reason of this decaying. Mechanisms of excitation and ionization processes of nitrogen molecules in this mixture have been studied. Processes which are responsible for the decaying of the population density of N₂(C³Π_u, ν') have been reported. Addition of H₂ to air improves the electrical characteristics of the DB discharge. An abrupt increasing in the electron density, reached about thirty fold at H₂ flow rate of 3 L/min, as a result of increasing the ionization processes has been reported. The breakdown voltage of the discharge decreased from 1.87 kV to about 1.25 kV by the addition of H₂ at flow rate of 3 L/min.

Keywords: Nitrogen Second Positive; DBD; H₂ Addition; Intensity Decaying

1. Introduction

It is known that the working conditions in the plasma may be affected, considerably, by the addition of small amounts of some molecular gases beside the working gas [1]. Recently, several experiments have been conducted already by various research groups, for studying the effects of adding of small amount of molecular gases in the discharge. The analytical parameters, such as the effective sputtering rate, the emission intensity of several spectral lines and the electrical current, have been affected by the addition of foreign gases to the operating gas. Mixed plasma gases may not always exert a positive influence on the analytical performance of the discharge characteristics, although it can be a possible option to improve the analytical performance [1].

The description of gas excitation in various plasma-chemical systems can be developed via the analysis of the population dynamics of reference levels for which rate constants of population by electron impact and following depopulation are well known. In the case of air DB discharge plasma, one such reference level is the C³Π_u state of N₂; its population determines radiation intensity of the 2⁺ nitrogen system transition

(C³Π_u → B³Π_g) in discharges with N₂. The short radiative lifetime (τ ≈ 40 ns) and relatively high excitation rate of this level make this transition suitable for diagnostics of the stationary plasma [2].

In the present paper the effect of addition of H₂ gas to air DB discharge on the optical and electrical characteristics is studied. These effects including the emission intensity of the nitrogen second positive systems (300 - 420 nm) and the relative population density of

N₂(C³Π_u, ν') states. Mechanisms of excitation and ionization processes of nitrogen molecules in this mixture are also studied.

2. Experimental Set Up

A DBD (dielectric barrier discharge) system, consists of two copper plane-parallel electrodes immersed in porous dielectric plates made of commercial gypsum (CaSO₄·2H₂O) material, has been used to produce an atmospheric pressure air discharge. The diameter of the dielectric plates was about 4 cm and its thickness was 2 mm. The distance between the two dielectric plates was kept constant at 1.1 mm. Hydrogen gas has been injected between the two electrodes at flow rates of (1 to 3) L/min.

The discharge open reactor was started up using a high voltage transformer (1 to 10 kV), generates sinusoidal voltage with frequency of 50 Hz. The applied potential (V_a), and the discharge current (I) were recorded using a digital oscilloscope (HAMEG HM407 - 40 MHz). The current was measured using a voltage drop across the resistance R_1 ($= 100 \Omega$) (see **Figure 1**).

An optical emission spectroscopy (OES) technique consists of a McPherson scanning monochromator [model 270] with a grating of 1200 grooves mm^{-1} and resolution of less than 2 \AA has been used to study the nitrogen spectra in a wavelength range of 300 - 420 nm. The monochromator was then connected to photomultiplier tube (PMT) type 9558 QB, which has a resolution time of less than 1 nanosecond, working at voltage of 1200 volts.

3. Results and Discussion

3.1. The Discharge Characteristics

Atmospheric pressure DB discharge can be operated in three different modes namely; the filamentary mode (streamers mode), the glow discharge mode (APGD mode) and the quasi-glow discharge mode. **Figure 2** represents the voltage-current waveform of the atmospheric pressure discharge in air at applied voltage of 3 kV and frequency of 50 Hz. Quasi-glow discharge mode has been generated whenever the dry gypsum porous dielectric material was used. The quasi-glow mode is characterized by the uniformity of the discharge current with only small filaments superimposed on the glow component [3-5].

3.2. Optical Emission Spectroscopic (OES) Characteristics

3.2.1. Effect of H_2 Addition on the Intensity of the Emitted Spectra

Typical emission spectra of the air DB discharge are shown in **Figures 3(a)-(d)** in the range of wavelength of (300 - 420 nm). **Figure 3(a)** shows the intensity of the nitrogen second positive systems in air discharge without admitting hydrogen gas. The second positive system is

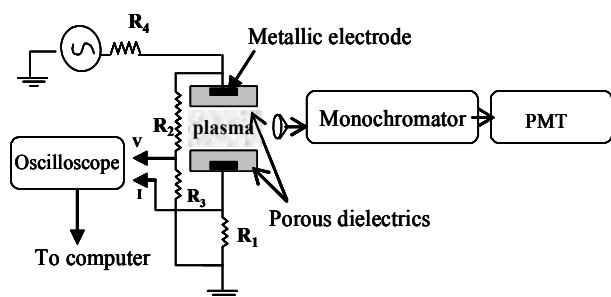


Figure 1. Schematic diagram of the discharge cell.

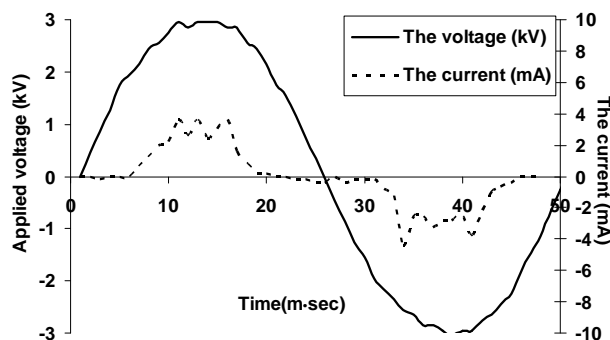
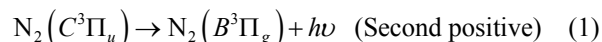


Figure 2. Voltage and current waveforms of air discharge using porous gypsum plates as barriers on the two electrodes, $d = 1.1 \text{ mm}$ and $V = 3 \text{ kV}$.

related to the de-excitation transitions of molecular nitrogen from the ($C^3\Pi_u$) excited electronic state ($v' = 0-2$) to the low-lying ($B^3\Pi_g$) excited state ($v' = 0-4$) according to: (Equation (1)), [6,7].



$B^3\Pi_g$ and $C^3\Pi_u$ states are lying at 7.4 and 11.0 eV respectively, above the ground electronic state. According to a simple analytical calculation for the electron mean energy in the atmospheric pressure, non-thermal plasma is estimated to be about (2 - 5) eV. Therefore the electronic states of the background molecules can be excited by the high-energy electrons in the tail of the Boltzmann distribution according to Equation (2), [8].

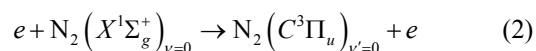
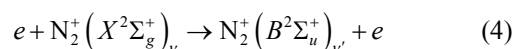
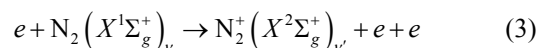
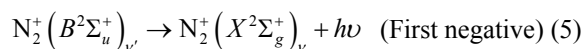


Figure 3(a) shows that a relatively large number of N_2 molecules are excited in the plasma, and hence strong emission spectra in the wavelength region of 300 - 400 nm are observed [8,9].

Figure 3(a) indicates that a weak intensity of (N_2^+)^{*} first negative transition is also observed. This transition is related to the radiative decay of the excited nitrogen ion (N_2^+)^{*}. The excited nitrogen ions results from the following two steps equations *i.e.* (3) and (4) [10]:



The de-excitation of (N_2^+)^{*} will followed by the emission of nitrogen First negative systems according to Equation (5), [10]:



Effect of addition of H_2 on the intensity of emitted spectra is shown in **Figures 3(b)-(d)**. It is noticed that:

1) There is an obvious decreasing in the intensity of the nitrogen second positive system by the addition of H_2

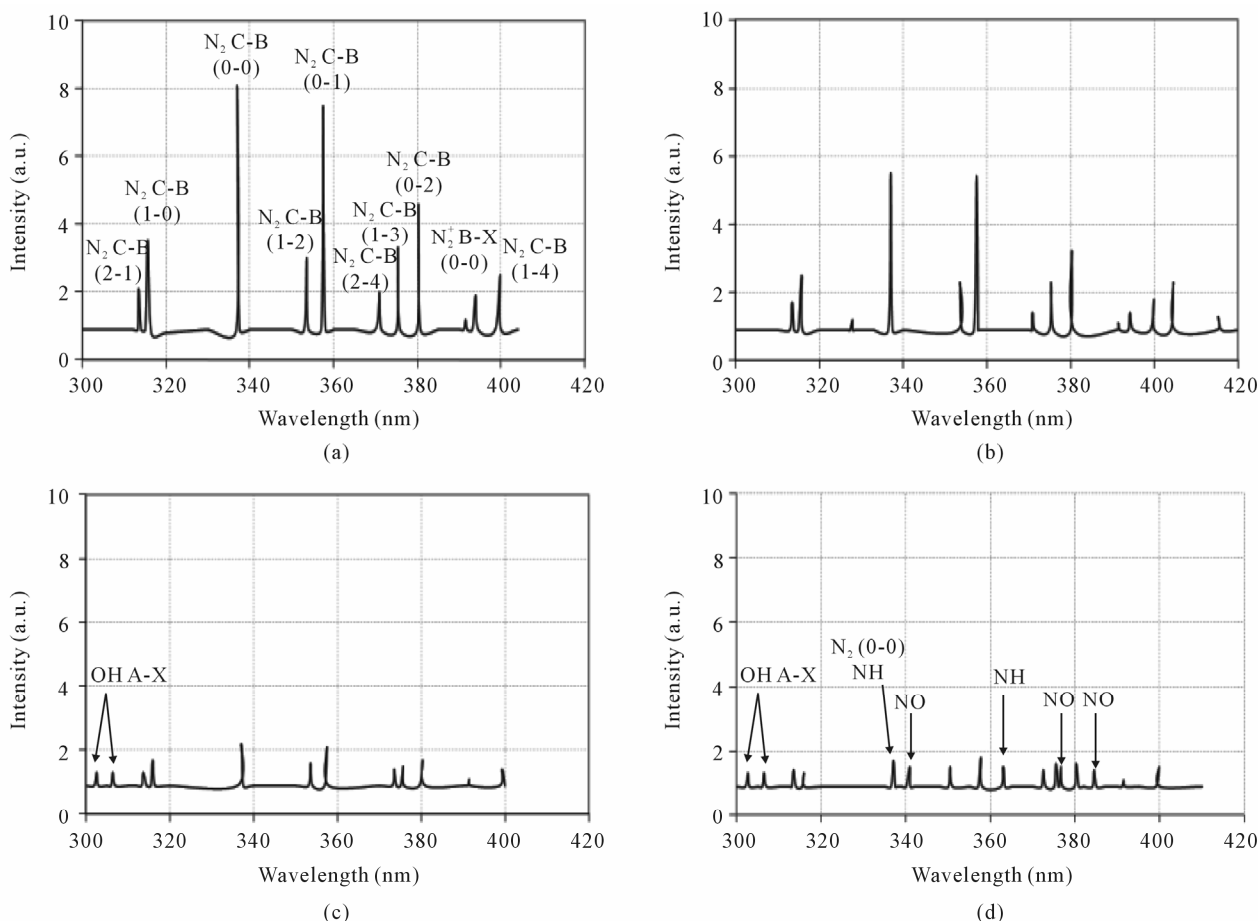
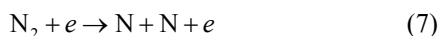
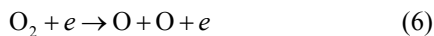


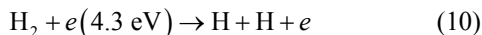
Figure 3. Typical emission spectra of air DB discharge in open air (a) and with addition of H₂ gas at flow rates 1 L/min (b), 2 L/min (c) and 3 L/min (d) where the discharge current is 1 mA.

gas, (see Section 3.2.2).

2) New species such as OH ($A^2\Sigma^+ - X^2\Pi_{3/2}$), NO and NH are observed in **Figures 3(c)** and **(d)**. This can be related to the fact that increasing of H₂ flow rate in air discharge gives rise to increase the dissociation processes of the nitrogen, hydrogen and oxygen molecules to form other species such as NO, OH and NH. NO radical is formed as a result of the dissociation of O₂ and N₂ by electron impact [11,12] according to Equations (6) and (7) and then re-combination of oxygen and nitrogen occurs (Equations (8) and (9)) [13]; *i.e.*



while the species NH and OH are formed according to the following equations;



then



3) No hydrogen lines are observed in **Figures 3(b)-(d)** e.g. H _{α} (at 656.2 nm), H _{β} (at 486.1 nm) or H _{γ} (at 434 nm). The disappearing of hydrogen lines is related to the exhausting of their excitation energy in the Penning ionization processes rather than the radiated decaying processes (see Section 3.3).

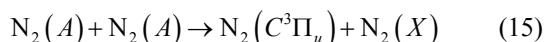
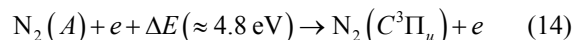
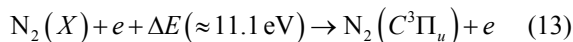
3.2.2. Effect of H₂ on the Decay of Nitrogen Second Positive System

The pronounced decay of the nitrogen second positive system ($C^3\Pi_u - B^3\Pi_g$) is attributed to:

1) Reducing the mean electron energy by the addition of hydrogen molecules to air discharge;

According to the fact that the dissociation energy of the hydrogen molecule (4.3 eV) is very low compared with that of the nitrogen (9.8 eV) or oxygen (5.11 eV) molecules, a considerable amount of electron energy is dissipated in the dissociation process of hydrogen molecules, Equation (10). As a result, the mean electron en-

ergy is reduced with the addition of H₂ to air discharge. Meanwhile, the production processes of N₂(C³Π_u) either by direct electron impact, Equations (13) and (14), or by the pooling reaction, Equation (15), is reduced.



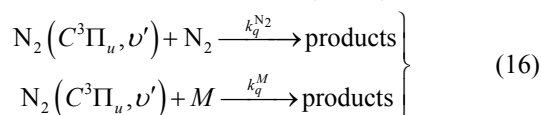
2) Reducing the formation of nitrogen excited molecules N₂(C³Π_u);

The dissociation processes of N₂ molecules increase by the increasing of H₂ to form another species such as NH and NO, etc. This in turns decreases the density of nitrogen molecules in the ground state that collide with the electrons to form the nitrogen excited molecules

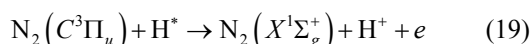
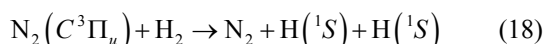
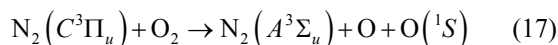
N₂(C³Π_u), (Equation (2)), that are responsible for the emission of nitrogen second positive system according to Equation (1).

3) Quenching of the formed N₂(C³Π_u) excited states before undergo the spontaneous emission to the low-lying N₂(B³Π_g) excited state;

This quenching is due to the collision of the nitrogen excited molecules N₂(C³Π_u) with N₂ or with other quenchers such as molecular hydrogen or oxygen respectively according to the collisional deactivation processes, Equation (16), [2] and its excited energy is exhausted in other reactions such as the dissociation of oxygen, Equation (17), [14] or hydrogen (H₂), Equation (18), [15] and [16], or excited hydrogen (H^{*}), Equation (19). The formed water (H₂O), by the re-combination of H₂ and O, is considered as a quencher for N₂(C³Π_u) [2]:



where $k_q^{N_2}$ is the quenching rate constant of N₂(C³Π_u) by N₂ and k_q^M is the quenching rate constant of N₂(C³Π_u) by M molecule, where M is O₂, H₂, H^{*} or H₂O.



The quenching mechanism of the nitrogen N₂(C³Π_u) is confirmed by using the Stern-Volmer Equation (20), [16,17] i.e.:

$$I_o/I_p = 1 + k_Q\tau[Q] \quad (20)$$

where I_o and I_p are the intensities of the special bands in the absence and the presence of the quencher

(which is H₂ in the present study), respectively, [Q] is the concentration of the quencher, τ is the lifetime of the nitrogen excited state, and k_Q is the quenching rate constant. In the present work, the hydrogen concentration can be represented by its flow rate in air. Therefore, by plotting the ratio $[(I_o/I_p)-1]/\tau$ vs the hydrogen flow rate, the quenching efficiency of the different bands of the nitrogen second positive system is estimated. **Figure 4** represents the Stern-Volmer plot of three selected bands of the nitrogen second positive system e.g. 313.6, 315.9 and 337.1 nm (life times τ 39×10^{-9} , 41×10^{-9} and 42×10^{-9} s) respectively [2].

The quenching rate of the band 337.1 nm (0 - 0) is the highest one (**Figure 4**), therefore, it is expected that the different quenchers prefer to react with this band rather than the other bands.

Figure 5 represents the effect of H₂ flow rate, in air DB discharge, on the intensity of the most popular nitrogen second positive band which (337.1 nm). A pronounced decaying of the intensity of this band by the increasing of H₂ flow rate is observed. The intensity of the NH band, at wavelength of 336 nm, is overlapped with that of the main band 337.1 nm.

3.2.3. Effect of H₂ Flow Rate on the Population Density of the Nitrogen Upper State N₂(C³Π_u)

In order to study the effect of addition of H₂ to air

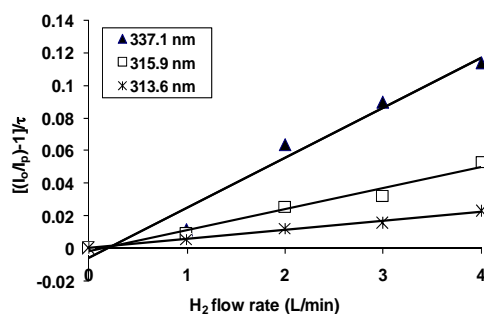


Figure 4. $[(I_o/I_p)-1]/\tau$ as a function of H₂ flow rates.

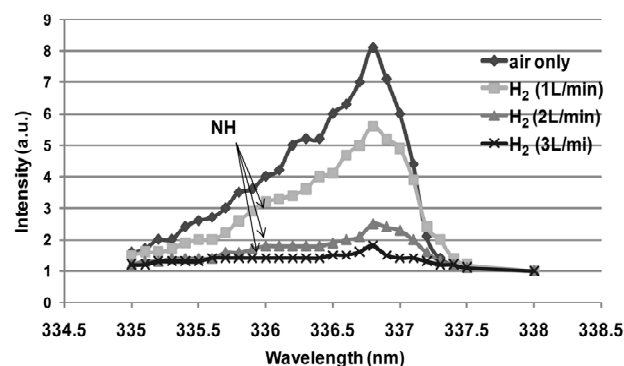


Figure 5. The emitted intensity of the nitrogen second positive band 337.1 nm at different H₂ flow rates.

discharge on the population density of the excited nitrogen states $N_2(C^3\Pi_u)$, Equation (21), which relates the intensity $I_{\nu'\nu''}$ of the $\nu'\nu''$ band of the system to the population density $N_{\nu'}$ of the excited state has been used [17] i.e.:

$$I_{\nu'\nu''} = DN_{\nu'}E_{\nu'\nu''}^4 R_e^2(\overline{r_{\nu'\nu''}})q_{\nu'\nu''} \quad (21)$$

where D is an instrumental constant; $E_{\nu'\nu''} = h\nu_{\nu'\nu''}$ is the quantum energy involved in the transition $\nu'\nu''$, ν is the frequency corresponding to the given band; $R_e(r)$ is the electronic transition moment; $q_{\nu'\nu''}$ is the Franck-Condon factor for the transition, both latter quantities are theoretically constant.

Figure 6 shows the normalized population density of $N_2(C^3\Pi_u)$, which is represented by the ratio

$$\frac{N_2(C^3\Pi_u)}{[N_2(C^3\Pi_u)]_{H_2=0}}$$

with and without addition of hydrogen to air discharge.

Figure 6 indicates a pronounced decay of the nitrogen population density of $N_2(C^3\Pi_u)$, by increasing the H_2 flow rate up to 2 L/min, then it saturates. The nitrogen population density decay confirms the quenching processes of the nitrogen $N_2(C^3\Pi_u)$ excited molecules (see Section 3.2.2).

3.3. Effect of H_2 Addition on the Electrical Characteristics of the Air Discharge

Figure 7 represents the voltage-current waveforms of the DB discharge using dry Gypsum dielectric material at hydrogen flow rates of 0, 2 and 3 L/min and using applied voltage of 3 kV at 50 Hz. Addition of hydrogen with different flow rates increases the peak current from (≈ 3 mA) without H_2 up to (≈ 13 mA) with H_2 at flow rate of 3 L/min.

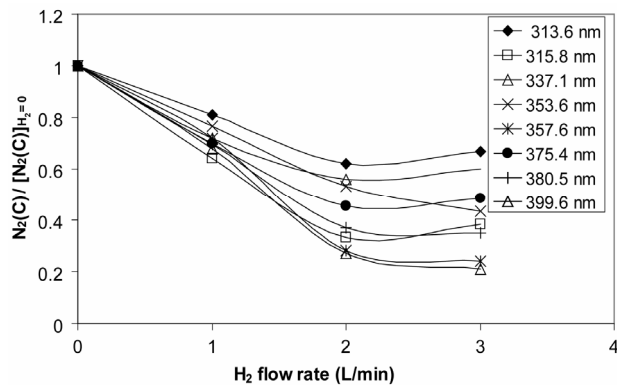
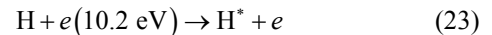
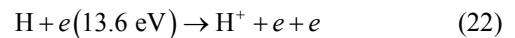


Figure 6. The normalized population density of nitrogen the upper state $C^3\Pi_u$ as a function of H_2 flow rates in air discharge.

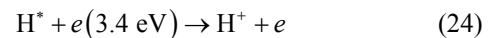
Figure 8 shows the peak values of the discharge current as a function of the applied voltage in air without and with addition of H_2 at different flow rates. Figure 8 confirms that increasing of the H_2 flow rate up to 3 L/min will increase the electric current by about 60% at the same voltage.

The enhancement of the discharge current by increasing the amount of H_2 is related to the following reasons;

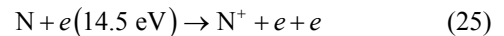
1) More ionization processes are expected to take place in the present of H_2 such as the reactions in Equations (19), (22)-(25) i.e.:



(H^* here is the hydrogen metastable state which has life time of 0.12 sec [25]) then



Also using Equation (7) then



2) Addition of H_2 to air discharge enhances the humidity of the porous dielectric that coated the electrodes.

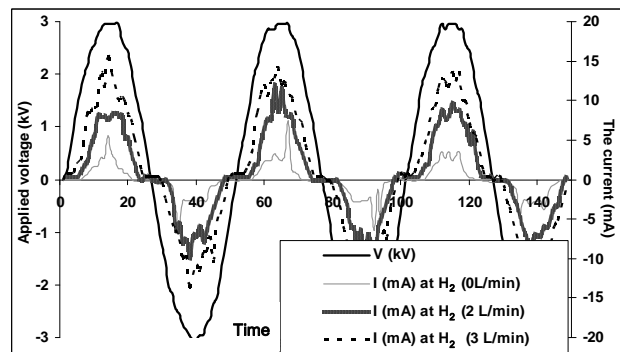


Figure 7. Voltage-current waveforms of air discharge in quasiglow mode at applied voltage 3 kV at different flow rates of hydrogen gas.

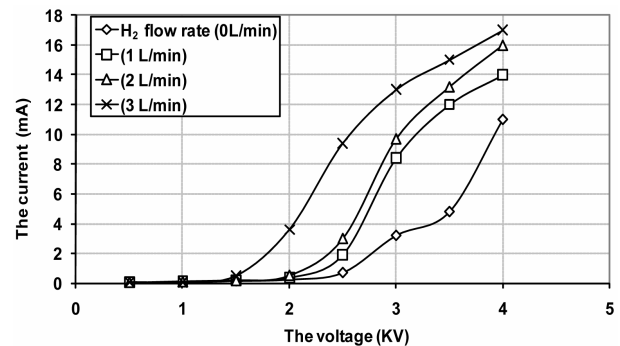
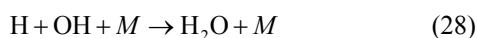
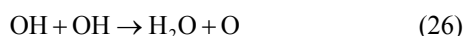


Figure 8. V-I characteristic curve of the DB discharge in air without and with H_2 at different flow rates.

This humidity resulted from the formation of OH and in turns promotes the formation of H₂O molecules according to Equations (26)-(28) [18] *i.e.*



Consequently, the humidity increases the current inside the micro-holes of the porous dielectric and hence increases the seed electrons that sustain the discharge in the glow mode [19].

3) Addition of H₂ to air discharge at atmospheric pressure increases the probability of Penning ionization and energy transfer processes. Addition of H₂ enhances also the dissociation processes of the molecular N₂ and O₂ (Equations (6) and (7)) to form large number of species, of different ionization and excitation energies. For example the product species NO, NH and O₂ have ionization energies; of 9.25, 13.1 and 12.07 eV [20] which can be ionized by another excited species of excitation energies higher than these values. The required energy can be obtained from the excited hydrogen or nitrogen states such as H(3d²D), H(4d²D), H(5d²D) and N₂(C³Π_u). The threshold excitation energies (from the ground state) for these states are 12.09, 12.75, 13.06 and 11.1 eV [21] respectively. The hydrogen excited atoms exhausted their energies in ionization processes rather than in radiative decaying processes. This explains the disappearing of the hydrogen lines

H_α(3d²D → 2p²P⁰), H_β(4d²D → 2p²P⁰) and H_γ(5d²D → 2p²P⁰) in the observed spectra (Figure 3(b)-(d)).

Figure 9 confirms a decreasing in the breakdown voltage by increasing of H₂ flow rate which is related to the increasing of the ionization processes by the addition of H₂ to air discharge.

3.4. Effect of H₂ Addition on the Electron Density in the Discharge

Figure 10 shows values of the electron density as a function of the hydrogen flow rate at applied voltage of 3 kV.

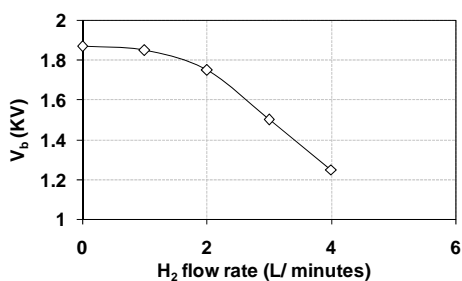


Figure 9. The breakdown voltage as a function of H₂ flow rate in air DB discharge.

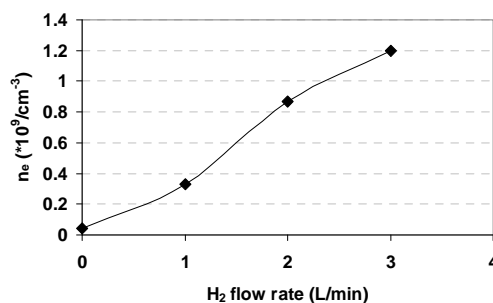


Figure 10. The electron density as a function of H₂ flow rates at applied voltage of 3 kV.

The electron density has been calculated using the electron conduction current density [22,23] *i.e.*:

$$J = -n_e e \mu_e E \quad (29)$$

where J is the discharge current density, μ_e is the electron mobility and E is the electric field of the discharge region.

Using the data in Figure 8, at applied voltage of 3 kV, the applied electric field E on the two electrodes is 2.7×10^4 V/cm. The electron mobility has been estimated by using Boltzmann solver, BOLSIG (KINEMA software) [24]. The software expresses the electron mobility as a function of E/P at a gas temperature of 300 K. In the present work $E/P = 35.52$ V·cm⁻¹·torr⁻¹, at this condition the mobility is found to be 5.13×10^2 cm²/V·sec. The electron density was estimated by substituting values of E , μ_e , and J in Equation (29).

Figure 10 indicates that addition of H₂ to air DB discharge causes an abrupt increasing in the electron density reached about thirty fold at flow rate of 3 L/min as a result of the mentioned increasing of the ionization processes.

3.5. Effect of H₂ Flow Rate on the Total Optical Emission Intensity of the Discharge

The total optical emission intensity of the discharge as a function of time has been measured using the PMT at different H₂ flow rates (= 0, 1, 2 and 3 L/min) in air discharge and at the same discharge current (~0.4 mA), see Figure 11.

The total emitted light decreases with the increasing of the H₂ flow rate. Addition of hydrogen to air DB discharge plasma enhances the ionization processes, such as Penning ionization, which occurs mainly by the energy transferring between the excited species. Since the present work has been carried out at atmospheric pressure the excited species will exhaust their energies through such ionization processes rather than in radiative decayed processes.

4. Conclusion

Admixing of hydrogen gas, with different flow rates, to

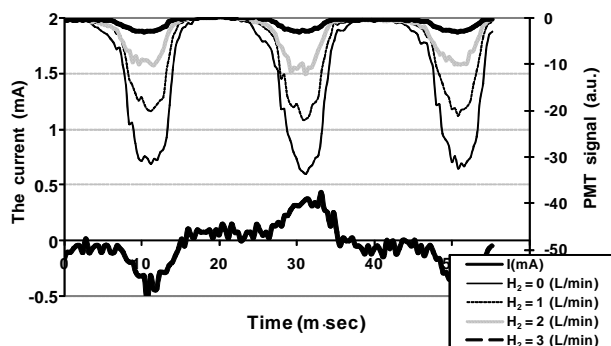


Figure 11. The Total optical emission intensity measured by a PMT and the discharge current as a function of time.

air DB discharge has an important effect on the optical and electrical characteristics of the discharge. An obvious decaying of the nitrogen second positive bands emission intensity was observed. This decaying of the nitrogen second positive bands resulted from the quenching of the nitrogen excited state $N_2(C^3\Pi_u)$. On the other hand, admixing the hydrogen to air discharge enhances the discharge current and in turns the electron density increases as a result of the increasing of the ionization processes e.g. the Penning ionization processes.

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