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Spectroscopic Analysis of DC-Nitrogen Plasma Produced using Copper Electrodes

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Abstract

This study shows the effects of copper material electrode, applied voltage, and different pressure values on electrical discharge plasma. The purpose of the work is the application of the spectral analysis method to obtain accurate results of nitrogen plasma parameters. By using the optical emission spectroscopy (OES), many N_2 molecular spectra peaks appeared in the range from 300 to 480 nm. Also, some additional peaks were recorded, corresponding to atomic and ionic lines for nitrogen, target material, and hydrogen, in all samples. The electron density (n_e) was calculated from the measurement of Stark broadening effect, which was found to decrease with increasing pressure from 0.1 mbar to 0.8 mbar. The higher emission intensities occurred at 0.2 mbar working pressure and were reduced with higher pressure. The vibrational temperature (T_{vib}) for N_2 increased from 0.17 to 0.33 eV with increasing the pressure from 0.15 mbar to 0.2 mbar, then decreased to 0.25 eV with increasing the pressure to 0.8 mbar. Other plasma parameters were studied, which are electron temperature (T_e), plasma frequency of electron (ω_p), and Debye length (λ_D).

Keywords:- Optical Emission Spectroscopy, Plasma Parameters, Glow discharge, Nitrogen plasma system.

التحليل الطيفي لبلازما النيتروجين ذو التيار المستمر المنتجة باستخدام أقطاب نحاسية

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الخلاصة

في هذا العمل تمت دراسة طيفية لتأثير أقطاب مادة النحاس مع الفولتية المسلطة والضغطوط المختلفة على بلازما التفريغ الكهربائي. اي ان الهدف من العمل هو طريقة التحليل الطيفي (OES) للحصول على نتائج دقيقة في دراسة بارامترات بلازما النيتروجين. حيث ظهرت العديد من قمم الأطياف الجزيئية N_2 في النطاق من 300 نانومتر إلى 480 نانومتر وبعض القمم الإضافية المقابلة للخطوط الذرية والأيونية للنيتروجين والمواد المستهدفة والهيدروجين ، في جميع العينات. تم حساب كثافة الإلكترونات للبلازما (n_e) والمحسوبة من تأثير توسيع ستارك ، حيث الكثافة الالكترونات تقل مع زيادة الضغط من 0.1 ملي بار الى 0.8 ملي بار .كذلك وجدت ان شدة الانبعاث الأعلى تكون عند ضغط تشغيل يبلغ 0.2 ملي بار. اما

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درجة حرارة الاهتزاز (T_{vib}) لجزيئات N_2 فتزداد من (0.17 إلى eV(0.33 مع زيادة الضغط من 0.15 ملي بار إلى 0.2 ملي بار ثم تنخفض إلى 0.25 eV مع زيادة الضغط إلى 0.8 ملي بار. معلمات البلازما الاخرى تم دراستها وهي تردد البلازما (ω_p) وطول ديباي (λ_D) ودرجة حرارة الالكترون(T_e)

Introduction

Glow discharge plasma is a region of relatively low temperature gas that is sustained in an ionized state by energetic electrons. The most commonly used method for the generation and sustainability of low temperature plasma for various technical applications is applying the electric field to neutral gases [1]. Any size of the neutral gas always contains a small number of electrons and ions that are formed as a result of the interaction of cosmic rays or radioactive radiation with gas. The method of spectral analysis of plasma light spectrum is used to diagnose and monitor plasma processes. From the emitted spectral lines, the spectral types can be determined by measuring intensity and wavelength that provide information about the chemical and physical operations in the plasma. The purpose of optical emission spectroscopy is to obtain as much information as possible, such as electron temperature (T_e) and electron density (n_e) within the limits of the plasma emission spectra in the optical range. It is a well-established non-invasive diagnostic technique with all necessary devices placed outside the system chamber, with proven cost and value effectiveness in basic and applied sciences [2, 3]. There are many studies on the generation and diagnosis of plasma in various fields [4-6]. The nitrogen plasma is generated for DC glow discharge system at different pressure and applied voltage values [7, 8]. The diagnostic tools used involve Langmuir probe and optical spectroscopy [9, 10].

The present work has two goals; the first goal is the study of the influence of work function on the N_2 plasma characteristics with different gas pressure. The second goal is the calculation of the vibrational temperature with DC applied voltage at copper electrode.

1- Theoretical Description

- The Boltzmann Equation for T_e

Optical methods employing spectral line emission intensity are widely used to measure internal plasma parameters (T_e and n_e) in the atmospheric pressure range. To calculate the electron temperature in the plasma, we can use the Boltzmann plot method. It is a simple and widely used method for OES measurements, based on the measurement of the relative intensities of two lines from the same element. However, in order to implement the Boltzmann plot method practically to measure the electron temperature, we must reach the excitation level under the specified conditions [11] .

The following equation allows to use the traditional technique to determine T_e using the Boltzmann relationship expression [12]

$$ln\left(\frac{\lambda_{ji}I_{ji}}{hcA_{ji}g_{ji}}\right)=\frac{-1}{kTe}\left(E_j\right)+ln\left(\frac{N}{U(T)}\right) \quad (1)$$

where I_{ji} are the relative intensity values of the emission line between the energy levels i and j , λ_{ji} is the wavelength, g_i is the statistical weight of the emitting upper level i of the studied transition, A_{ji} is the transition probability for spontaneous radiative emission from the level i to the lower level j, E_j is the energy of excitation at level i, k is the Boltzmann constant, and N is the number of densities.

- Stark Brooding for n_e

The electron number density is an important parameter used to describe the plasma environment and is crucial for establishing its equilibrium status. The electron number density is usually measured from the Stark broadening, where the electron density can be determined from the line width by using the following equation [13]:

$$n_e(cm^{-3}) \;=\left(\frac{\Delta\lambda}{2\omega_s\,(\lambda Te)}\right)N_r \quad (2)$$

where $\Delta\lambda$ is the full width at half maximum (FWHM) of the line, ω_s is the Stark broadening parameter which can be found in the standard tables, and N_r is the reference electron density which is equal to 10^{16} cm^{-3} for neutral atoms and 10^{17} cm^{-3} for single charged ions.

- Plasma frequency (ω_{pe})

Plasma frequency of electron (ω_{pe}) can be calculated as [14]:

$$\omega_{pe}=\sqrt{\frac{e^2 n_e}{\varepsilon_o m_e}} \tag{3}$$

where, ε_o is the permittivity of free space, n_e is electron density, e electron charge, and m_e is electron mass.

- Debye length (λ_D)

Debye length is an important parameter in plasma. If an electric field is created in the plasma, then the charged particles will interact to reduce the field effect. Debye shielding gives the plasma its quasi-neutrality characteristic.
The Debye length is defined by the following equation [15]:

$$\lambda_D = \left(\frac{k\, T_e\, \varepsilon_o}{n_e\, e^2}\right)^{1/2} \tag{4}$$

2- Experimental setup

This work contains details of various experimental techniques and devices, such as the DC discharge system (i.e. vacuum system, vacuum chamber, power supply and spectrometer) and the experimental procedures used to study the discharge plasma characteristics in nitrogen at low pressure. Figure 1 shows the schematic diagram of the experimental work procedures that were achieved in this work. The vacuum chamber is made of glass with a diameter of 30 cm and a height of 40 cm, with a transparent plastic base with a rubber ring, gas entrance, and electrical feed-through.

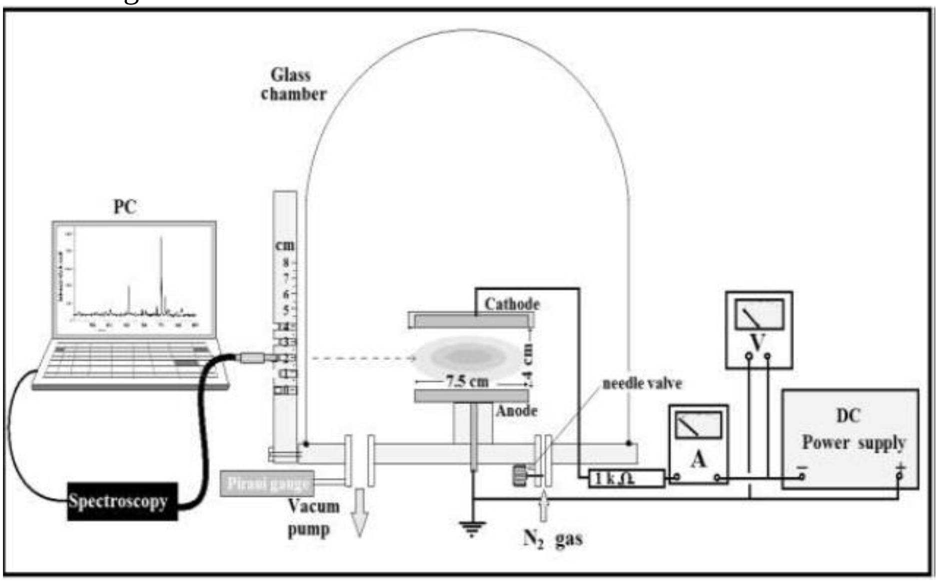

Figure 1- Schematic chamber for studying N_2 gas discharge characteristics

The atmospheric gases were physic-chemisorbed on the interior surfaces, arising from the exposure of these surfaces to ambient atmosphere. Thus, the first step in this procedure was the degassing of the rotary pump (backing) to reduce the pressure inside it and to eliminate the heavy molecules that prevent increased vacuum. This process continued for a period of 15 minutes. After that, the chamber was vacuumed down to 5×10^{-3} Torr. At the end of these procedures, we started to deliver the nitrogen gas inside the chamber with purity of 99.9%, until reaching atmospheric pressure, to ensure the presence of only the nitrogen gas molecules inside the chamber. Then, the chamber was evacuated to selected pressure values.

Figure 2 shows a photograph of the main experimental setup of DC discharge plasma system used in this work, which consisted of double stage rotary pumps, glass chamber, Perini head and reader, H.V DC-power supply, multi-meters for discharging current measurements, high voltage voltmeter, and gas source. Gas source - flow control system that was used to deliver the feed to the plasma chamber with the required flow rate and gas pressure. It consisted of nitrogen gas storage cylinders, regulators, tubes, and a needle valve.

Figure 2- Illustration of DC glow discharge system

After the configuration of the system and the measuring instruments attached to it, we started the experiment which included several steps. The atmospheric gases physic-chemisorbed on the interior surfaces arise from the exposure of these surfaces to ambient atmosphere.

The first step in this procedure was the degassing of rotary pump (backing) to reduce the pressure inside this pump and to eliminate the heavy molecules that prevent increased vacuum. This process continued for a period of 60 minutes. After that, the chamber was vacuumed down to 5×10^{-3} Torr.

Results and Discussion

Spectroscopic measurements were performed for the glow produced by DC discharge plasma in nitrogen gas. Planner electrodes with copper target material were used at different working pressure, from 0.15mbar to 0.8 mbar, and constant electrodes spacing of 3 cm. The effects of the used electrode, applied voltage, and working pressure on plasma parameters were studied by comparing the produced lines with neutral and ionic standard lines for the used target material; molecular, atomic, and ionic lines for nitrogen gas.

Plasma emission spectroscopy was performed using copper electrode with different pressure values. Figure 3 shows the spectroscopic patterns of the emission from DC plasma in nitrogen gas with 3 cm inter-electrode distance at constant applied voltages of 100V and different working pressures (0.15, 0.2, 0.3, 0.4, and 0.8 mbar). These patterns were compared with those of N_2 molecular lines and atomic and ionic lines corresponding to nitrogen, copper, and hydrogen atoms. The N_2 molecular peaks were limited in the range from 300 to 480 nm, which dominated this region. In addition, several peaks corresponding to atomic and ionic lines for copper, nitrogen, and hydrogen were recorded in all samples. One can observe that the peaks of intensity increased gradually by increasing working pressure from 0.15 to 0.2

mbar, as a result of increasing excited and ionized atoms or molecules with increasing the molecular density with pressure. Then, the peaks decreased with increasing pressure to 0.8 mbar, due to the reduction in the mean free path for electrons, which prohibited the electrons from gaining the required energy between two excitation or ionization collisions. As mentioned before, a hydrogen line appeared as a result of the presence of residual water vapor desorbed from discharge chamber walls.

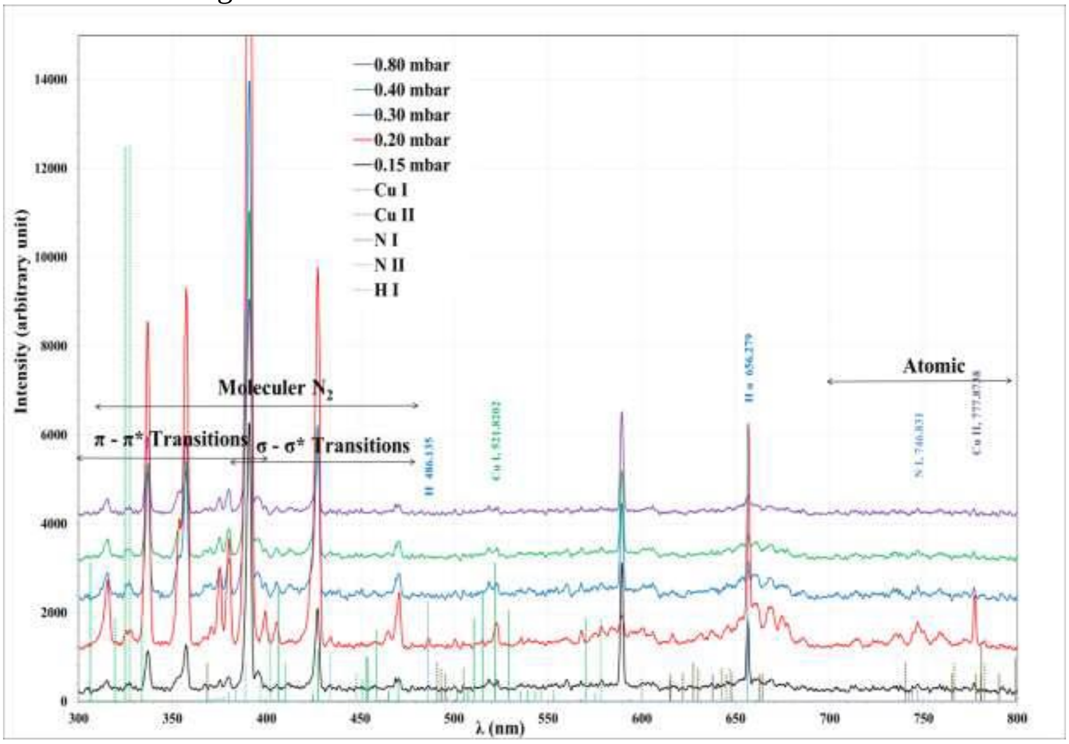

Figure 3 -Plasma emission patterns produced by DC discharge in nitrogen using copper target with different working pressure values.

The values of electron temperature were calculated by the ratio method, using the intensities of the two hydrogen lines (656.28 nm and 486.13 nm) for different working pressure values. While electron density was calculated by Stark broadening effect, using the line broadening (Δλ) for Ha line with different working pressure values, as shown in Figure 4. The full width at half maximum for all curves was found by Gaussian fitting. The standard values of broadening for this line were employed to calculate electron density, which is directly proportional to Δλ. It can be seen that the full width increased with increasing the pressure from 0.15 to 0.4 mbar, then reduced at 0.8 mbar.

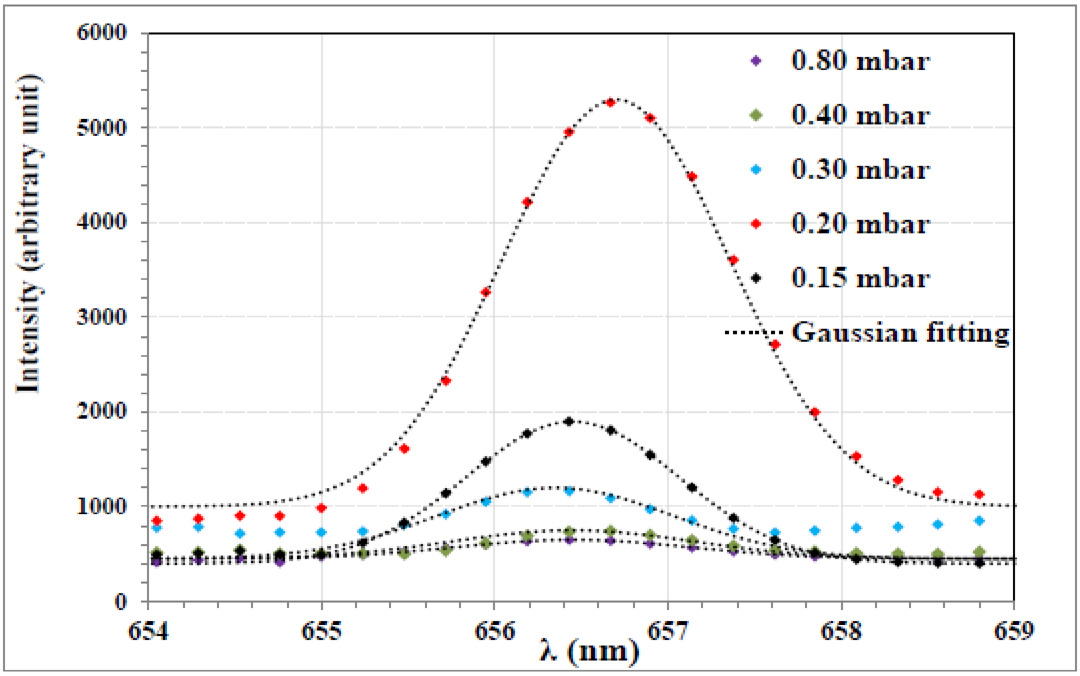

Figure 4-Hα (656.28 nm) peaks broadening and Gaussian fitting at different working pressure values using Cu electrode in nitrogen gas**.**

Figure 5 illustrates the variation of electron temperature and electron density for DC discharge of nitrogen gas at different working pressure values. This figure shows that T_e decreases from 0.386 to 0.364 eV with increasing pressure from 0.15 to 0.4 mbar, then increased to 0.376 eV at 0.8 mbar due to increasing the energy transfer from electrons to atoms by excitation and ionization collision, which increased with increasing density, then reduced with higher density. The electron density had an opposite behavior, being increased from 7.25×10^{16} to 8.25×10^{16} cm^{-3} with increasing pressure from 0.15 to 0.4 mbar, then decreased to 8.0×10^{16} cm^{-3} at 0.8 mbar.

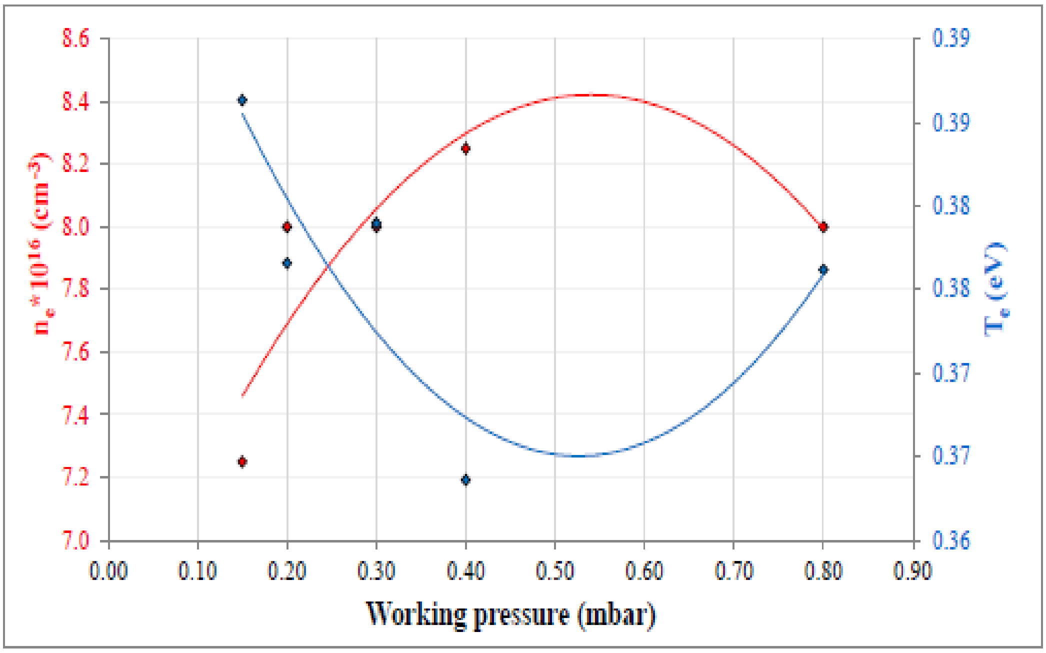

Figure 5- The variation of T_e and ne for DC discharge in N_2 with working pressure using Cu electrode at constant applied voltage (1100 V) with 3 cm inter-electrode distance.

Table 1 shows the calculated values of λ_D, ω_p, and n_e for DC plasma in N_2 gas with different working pressure values, using Cu electrode.

Table 1- Plasma parameters for DC discharge N_2 gas with different working pressures using Cu electrode**.**

P (mbar)	T_e (eV)	n_e x10^{16} (cm^{-3})	ω_p x10^{12} (rad/s)	λ_D x 10^{-6} (cm)
0.15	0.386	7.250	15.185	3.246
0.20	0.377	8.000	15.951	3.223
0.30	0.379	8.253	15.952	3.156
0.40	0.364	8.255	16.198	3.158
0.80	0.376	8.000	15.951	3.155

The molecular transitions appeared between 320 nm to 480 nm for samples with different working pressure values (0.15 to 0.8 mbar), as shown in Figure 6. The peaks corresponding to π- π^* transitions, indicated with red color, and to σ –σ^* transitions, indicated with blue color, are for nitrogen molecular peaks. The energy difference in these transitions comes from the summation of electronic, vibrational, and rotational energies. The peaks indicated with π- π^* for pure electronic transition (Δv=0) are located at 336.6 nm for π- $\pi^*$and at 391.4 nm for σ –σ^*. The other lines corresponding to Δv values that are not equal to zero are shifted, with energy values equal to the multiple of energy difference between the vibrational levels and rotational energies. It can been noticed that the intensity of emission peaks increased with increasing working pressure from 0.15 to 0.2 mbar, due to increasing the density of the excited molecules, then reduced with higher pressure.

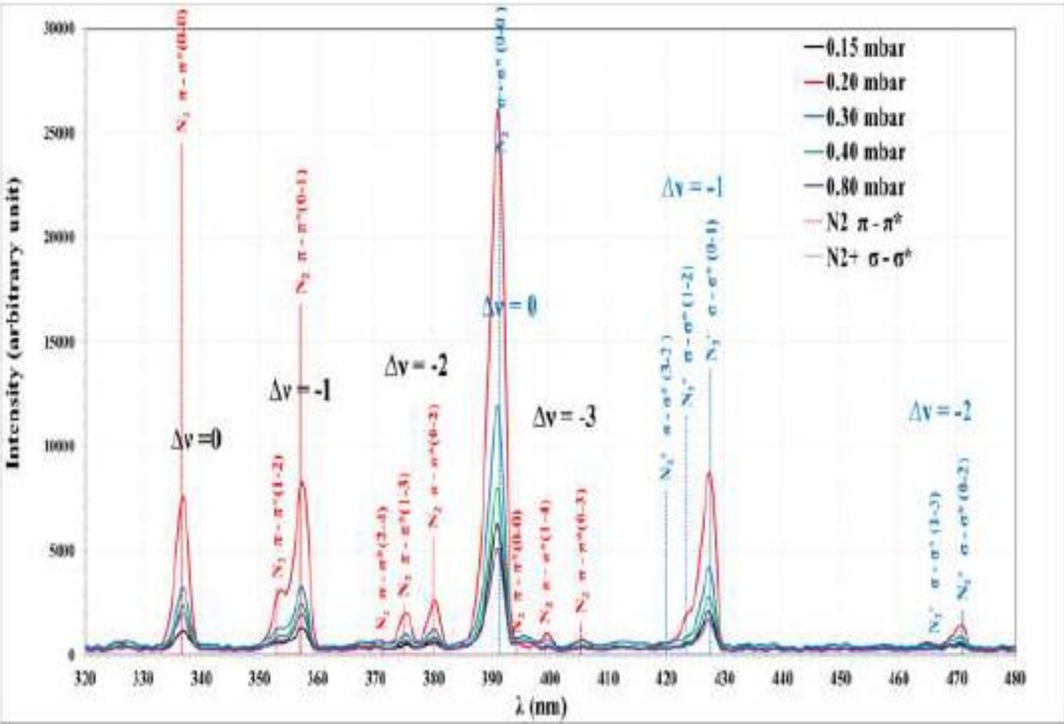

Figure 6-The results of matching the emission patterns, for different working pressures using copper electrode, with Nz molecular lines.

Figure-7 shows the variation of line intensities for the pure electronic transitions in the nitrogen molecules (λ= 336.6 nm and 391.4 nm). All peak intensities for 391.4 nm are higher than those for 336.6 nm. The higher intensities were found at 0.2 mbar working pressure, which were reduced with higher pressure.

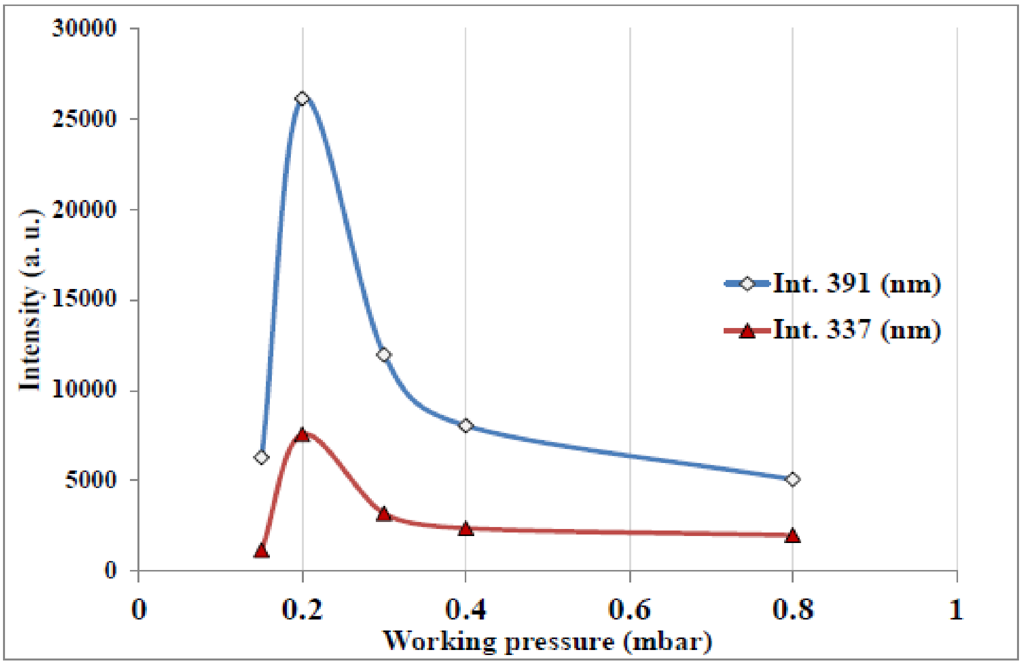

Figure 7-Variation of line intensities for pure electronic transition with working pressure using copper electrode.

Figure 8 shows the variation of intensity ratio between the two lines with working pressure. The intensity ratio increased at 0.2 mbar, due to increasing the excited collisions at this pressure, then decreased with higher pressure, due to reducing temperature.

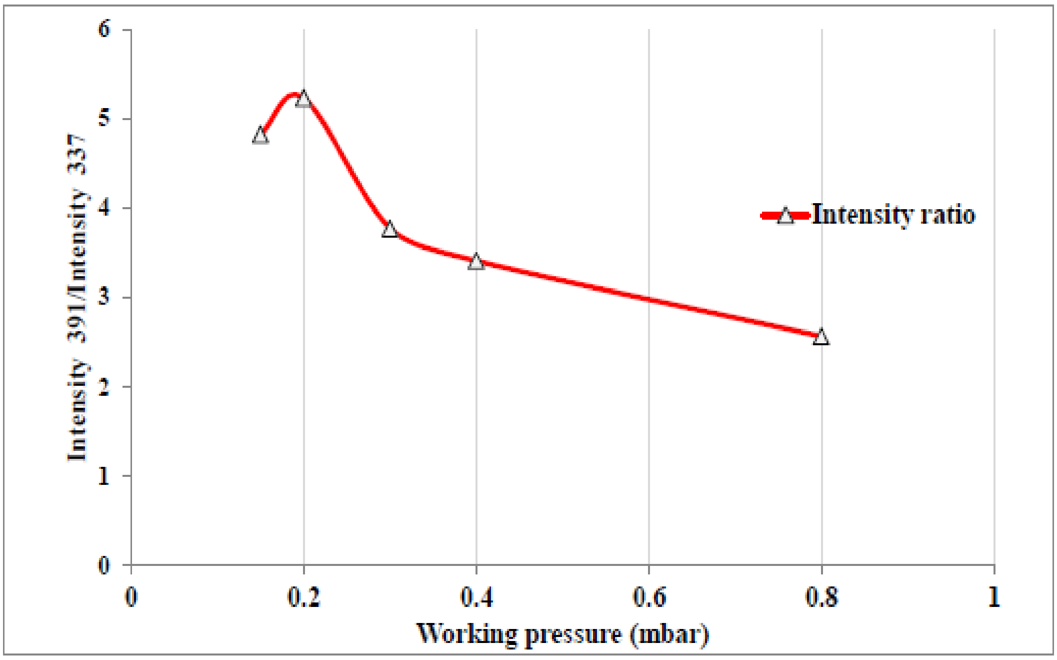

Figure 8- Variation in the intensity ratio of the two lines ($I_{391.4}$:$I_{336.6}$) with working pressure using copper electrode.

The vibrational temperature for N_2 molecular lines, for emission produced by DC discharge at different pressure values, was calculated by employing the lines with Δv = -2 for the transitions from the vibrational quantum numbers of 0-2, 1-3 and 2-4. For this purpose, Spectrum Analyzer 1.7 software was utilized, using the probability of transitions and energy levels for all patterns with different working pressures. Figure 9 displays the variation of vibrational temperature (T_{vib}) with working pressure. The results show that T_{vib} increased from 0.17 to 0.33 eV with increasing the pressure from 0.15 to 0.2 mbar, then decreased to 0.25 with increasing the pressure to 0.8 mbar.

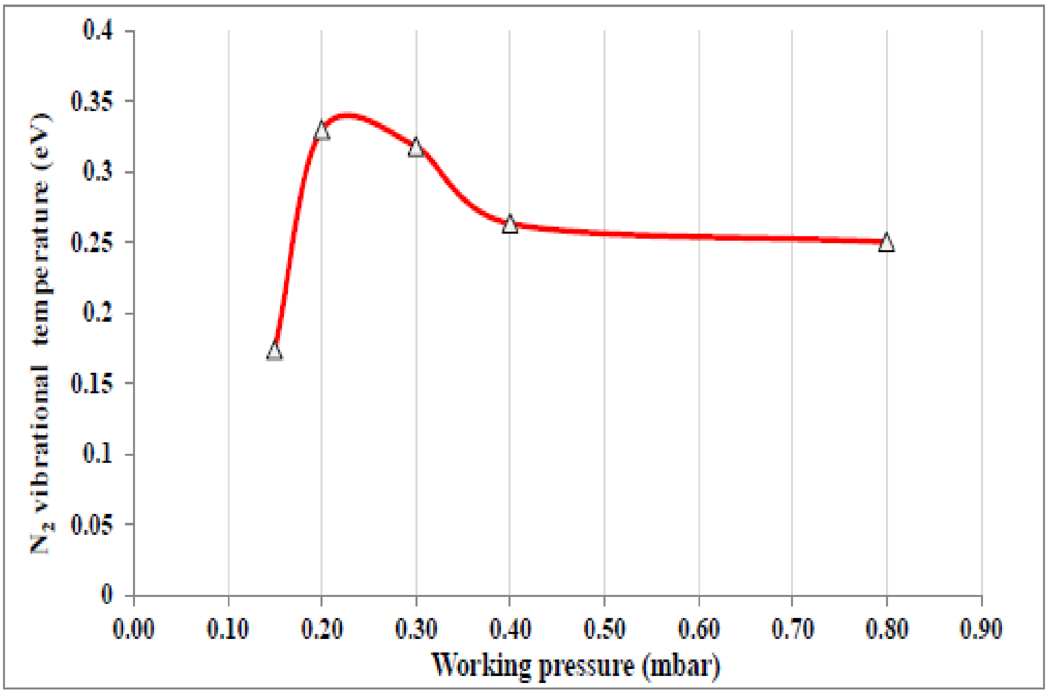

Figure 9-Variation of vibrational temperature (T_{vib}) as a function of working pressure

3- Conclusions

Several main conclusions could be obtained from this research.

There are many N_2 molecular peaks in the range from 300 nm to 480 nm and some additional peaks corresponding to atomic and ionic lines for nitrogen, electrodes material, and hydrogen in all samples. The electron temperature decreases with increasing pressure from 0.15 to 0.4 mbar due to increasing the energy transfer from electrons to atoms by excitation and ionization collision, then increases as a result of decreasing the energy delivered to electrons. This decrease leads to a decrease in the plasma density due to decreasing the ionization collision's cross section.

The vibrational temperature for N_2 molecular is lower than the calculated electron temperature. While T_{vib} increases with increasing the pressure from 0.15 to 0.2 mbar, then decreases with increasing the pressure to 0.8 mbar.

The higher emission intensities occur at 0.2 mbar working pressure, due to increasing the excited collisions at this pressure. Then, they decrease with higher pressure for the copper electrode due to reducing temperature.

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