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Spectroscopic Diagnosis of Cobalt Plasma Produced by OES Technique and Influence of Applied Voltage on Plasma Parameters

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Abstract

In this work, plasma system that operates at vacuum was designed and built using a sheet of cobalt metal for the purpose of diagnosing plasma and measuring its parameters, as it is very important to know the processes that accompany plasma generation and are closely related to them, including the electron density in the plasma and its temperature. The spectroscopic diagnosis was done by optical emission spectroscopy (OES) which relies on the calculation of the optical radiation emitted by the plasma to describe plasma parameters in the chemical, molecular, and ionic radiator's near environment, and applied to cobalt metal at vacuum D.C high voltage power supply. The results showed the rise of spectral lines intensity with increasing the applied voltage. The maximum peak of argon gas (ArI) was at the wavelength (811.5311) nm and the maximum peak of cobalt metal (CoI) was at the wavelength of 242 nm, where argon gas was used at the fifth flow per minute with variable voltages (13-21) kV. The results also achieved that the values of electron temperature rises from (0.2708-0.6649) eV with the increase in the applied voltage, as well as the electron density from (8.108-13.851)x$10^{17}$$cm^{-3}$ with the stability of the argon gas flow rate at 5 *l/min*. The length of the plasma was measured at different gas flow rates (1-5) *l/min* and different applied voltages (13-21) kV that were used in this diagnosis.

Keywords: Cobalt Plasma Jet, Emission Spectroscopy, Plasma Parameters, Non-Thermal Plasma, Plasma Length.

التشخيص الطيفي لبلازما الكوبالت الناتجة باستخدام تقنية OES وتأثير الفولتية المسلطة على معلمات البلازما

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الخلاصة

في هذا العمل، تم تصميم وبناء منظومة بلازما تعمل عند الضغط الجوي واستخدام شريحة من معدن الكوبالت لغرض تشخيص البلازما وقياس خصائصها، حيث من المهم للغاية معرفة المعلمات المصاحبة لتوليد البلازما والتي ترتبط ارتباطًا وثيقًا بها، بما في ذلك كثافة الإلكترون في البلازما ودرجة حرارته، حيث تم بناء هذه المنظومة وتطبيق التشخيص الطيفي البصري الذي يعتمد على حساب الإشعاع البصري المنبعث من البلازما لوصف معلمات البلازما في البيئة القريبة من خلال الشعاع الكيميائي والجزيئي والأيوني الصادر من

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معدن الكوبالت عند الضغط الجوي مع فرق جهد عالي مستمر، أظهرت النتائج ارتفاع شدة الخطوط الطيفية مع زيادة الجهد او الفولتية المسلطة، و كانت أقصى قمة لغاز الأرجون (ArI) عند الطول الموجي (811.5311) نانومتر والقمة القصوى لمعدن الكوبالت (CoI) كان عند الطول الموجي 242 نانومتر، حيث تم استخدام غاز الأرجون عند التدفق الخامس لكل دقيقة مع فولتية متغيرة (13−21) كيلو فولت، كما تم تحقيق ارتفاع في قيم درجة حرارة الإلكترون مع زيادة الفولتية المسلطة من (0.2708−0.6649) الكترون فولت وكذلك كثافة الإلكترون من (8.108−13.851) × 10^{17}سم $^{-3}$ مع ثبات تدفق غاز الأرجون 5 لتر/دقيقة، تم قياس طول البلازما لتدفقات مختلفة من الفولتية التي استخدمت في هذا التشخيص.

1. Introduction:

The use of atmospheric pressure plasmas in a wide range of applications in the biological, nanotechnology, and agricultural fields has recently piqued interest, this is particularly true in the biomedical field, where applications include bacteria inhibition, tissue regeneration, and dental bleaching. Atmospheric plasmas have also attracted attention in industry, with applications such as enhancing adhesion for inks, paints, and coatings being examples of plasma uses in order to improve surface qualities[1-4]. Due to the higher temperature of electrons in these plasmas compared to heavy particles, even though elastic collisions of electrons are less effective than collisions of heavy particles, they can still transfer energy to other processes such as ionization, activation, or dissociation of molecules, which explains the high interest in these plasmas[2-3]. Plasma has free charging particles at the microscopic level, with both negative and positive charges storing roughly the same amount of energy[4]. This occurs when the elements that make up a certain state of matter are heated to a temperature higher than the thermal energy but lower than the binding energies of those elements [5]. The goal of a plasma diagnostic such as electrical sampling and optical spectroscopy of emissions (OES) is to obtain information about plasma parameters through the use of a variety of experimental techniques [6].To determine plasma attributes, it is necessary to understand the effects of the numerous physical processes that occur and to determine the effects of these processes[7]. Spectroscopic procedures, such as laser dispersion, emission, absorption, and fluorescence spectrometry, are unique methods for collecting distinct sections of plasma without altering its status or structure[8]. One of these techniques is optical emission spectroscopy (OES), which relies on the calculation of the optical radiation emitted by the plasma to describe plasma parameters in the chemical, molecular, and ionic radiator's near environment[9]. So, in order to obtain information about plasma, spectroscopic diagnosis was used to calculate and measure these parameters, such as electron density, excited species state densities, collisional electron–atom, atom–atom, and ion–atom effects, energetic distribution of species, temperature of species, charge transfer between plasma components, rotating structure of molecules, and even electric charge. Spectroscopy is a non-intrusive method of analyzing electromagnetic radiation from a plasma source; atomic spectra are primarily concerned with the exchange of energy between the atom and electromagnetic radiation, which can be associated with a valence electron changing its orbit in the simplest model[10]. Energy can be added to the radiation field (emission spectra) or withdrawn from it (absorption spectra), the frequency of the radiation absorbed or emitted is related to the actual change in energy (ΔE) between the energy levels of an atom by the equation[1]:

$$E' - E'' = \Delta E = h\nu \quad (1)$$

Where: E' is the total energy of the atom in its higher state and E'' is the total energy of the atom in its lowest state, h is Planck's constant and ν is the frequency. Non-equilibrium plasmas can be created under atmospheric pressure discharge circumstances, with the majority of them being easily handled. These discharge plasmas can be created in a liquid state, which opens up a wider range of applications, such as in the medical, food, agricultural, and textile industries[11]. The Scientist McWhirter proposed four plasma models based on electron interaction mechanisms, local-thermal-equilibrium (LTE), stable-state corona, time-dependent corona, and collision radiative models [12]. Collisional excitation is balanced in low-density plasmas by spontaneous decay, and collisional ionization is balanced by radiative recombination. For such low-density plasmas, the population densities of the new rates are computed using a balance between radiative decay to ground state and collisional excitations. the temperature of the plasma is a crucial thermodynamic attribute because of its ability to recognize and forecast particular properties of plasma, such as populations with particle velocity distribution and relative energy levels[13]. The electron temperature (T_e eV) for LTE is calculated using the following equation[14]:

$$ln\left[\frac{\lambda_{ji} I_{ji}}{hcA_{ji} g_j}\right] = -\frac{1}{KT}\left(E_j\right) + \ln\left|\frac{N}{U(T)}\right| \quad (2)$$

Where: g denotes the statistical weight, λ denotes the wavelength, E is the excited state energy in eV, I_{ji} denotes the intensity, A_{ji} denotes the transition probability, N denotes the density of the state's population, and k denotes the Boltzmann constant. There are a number of viable ways for estimating electron density, including plasma spectroscopy, laser interferometry, Thomson scattering, and microwave[14]. A well-known method for determining electron density employs a linear Stark spectral line extension. The impact of Stark and Doppler's breadth is responsible for broadening the line in a plasma jet; the latter is solely dependent on the temperature and atomic mass of the emitting species[15]. The Stark effect, which encompasses interactions for both surrounding particles and radiators, can broaden the pressure; ion collisions between fewer electrons produce these interactions. The Stark broadening is proportional to the local electric fields created by adjacent charge particles, notably electrons around emitting atoms, as a function of their density and energy, which makes measuring electron density and field strength easier[3]. Consequently, Stark broadening is one of the most essential spectral line widening aspects in plasma spectrum analysis because it offers electron density at the specified plasma electron temperature[6]. The Gaussian profile is typically produced by instrumental broadening, natural broadening; Doppler broadening and Stark broadening can lead to a Lorentzian profile if the electron impact widening is taken into account while the ion dynamics are ignored during the radiative process. As a result, the electron density can be calculated using the following formula [14]:

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

Where: Δλ is the full width at half maximum (FWHM) nm of the line, and ω_s is the Stark broadening parameter that can be found in the standard tables, N_r is the reference electron density. The frequency of plasma is determined from the following equation[13]:

$$f_p = 8.98\sqrt{n_e} \ (\text{Hz}) \quad (4)$$

One of the most fundamental properties of plasma is its frequency, which is solely dependent on density. Because electrons have such a small mass, plasma has a high frequency. Debye shielding λ_D is a charged particle response that reduces the impact of electric on local fields, giving the plasma its quasi-neutrality. Debye length(λ_D)is given as[14]:

$$\lambda_D = \sqrt{\frac{\varepsilon_o K_B T_e}{n_e e^2}} = 743 \times \sqrt{\frac{T_e}{n_e}} \quad (cm) \tag{5}$$

The first condition for plasma formation is that the Debye length should be as short as possible in relation to the system dimension, to examine the formation of active species[16].
This research describes argon plasma in terms of argon percent and operational parameters using OES. The Boltzmann plot method was used to estimate the electron temperature, which also effects plasma reactivity through inelastic collisions that produce the active species. Understanding the mechanisms that govern the development of plasma reactivity species is one of the main objectives of this paper.

2. Experimental Setup

A plasma system that operates at atmospheric pressure was designed and built. This system used a power supply (working in two directions: continuous voltage and alternating voltage) with maximum voltage of 25 kV and a cut-off frequency of up to 30 kHz. Tubes were used to transfer argon gas from the cylinder (Argon bottle) to the jetting needle through a gas control unit included in specific divisions of gas flow ranging from 1 to 5 liters per minute of gas. The plasma needle with an outer diameter of 3 mm was designed with a stainless steel tip. The head of the plasma jet was connected to the cathode electrode of the high voltage power supply, and the other electrode of the high voltage power supply (anode) was connected to the cobalt metal (Co) below the head of the plasma jet in the beaker as illustrated in Figure 1. A spectrometer (S3000-UV-NIR) was connected with the system for the purpose of directly recording the diagnostic data at different gas flows as shown in Figure 1. The spectra were recorded in the range (160-1010) nm directly from the plasma jet, where the plasma spectrum of argon gas was obtained at the wavelength of (700-850) nm, as well as with the cobalt spectrum data (CoI) recorded at the wavelength of (231-255) nm. These recorded data were analyzed and matched with the data of the National Institute of Technology and Standards (NIST). Finally, the plasma parameters of cobalt metal were extracted and its properties were discussed.

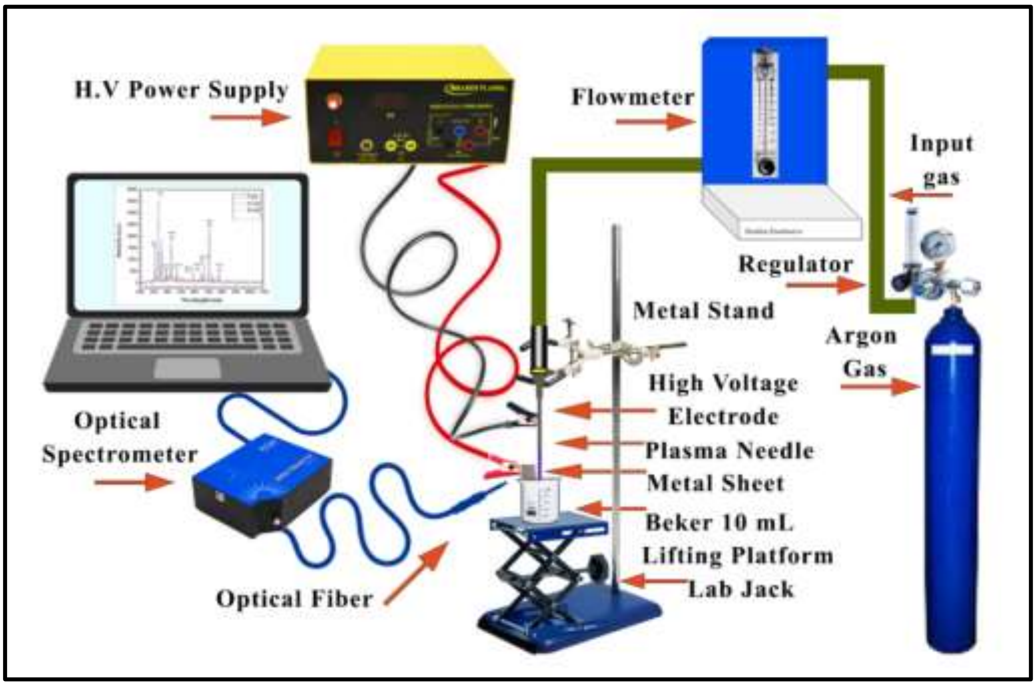

Figure 1: Schematic diagram of the atmospheric cobalt plasma jet system.

3. Results and Discussion

Figure 2 shows the intensity distribution of plasma spectrum obtained using OES at applied voltage in the range (13-21) kV with frequency of 6 kHz, for argon gas flow rate of 5 *l/min.*

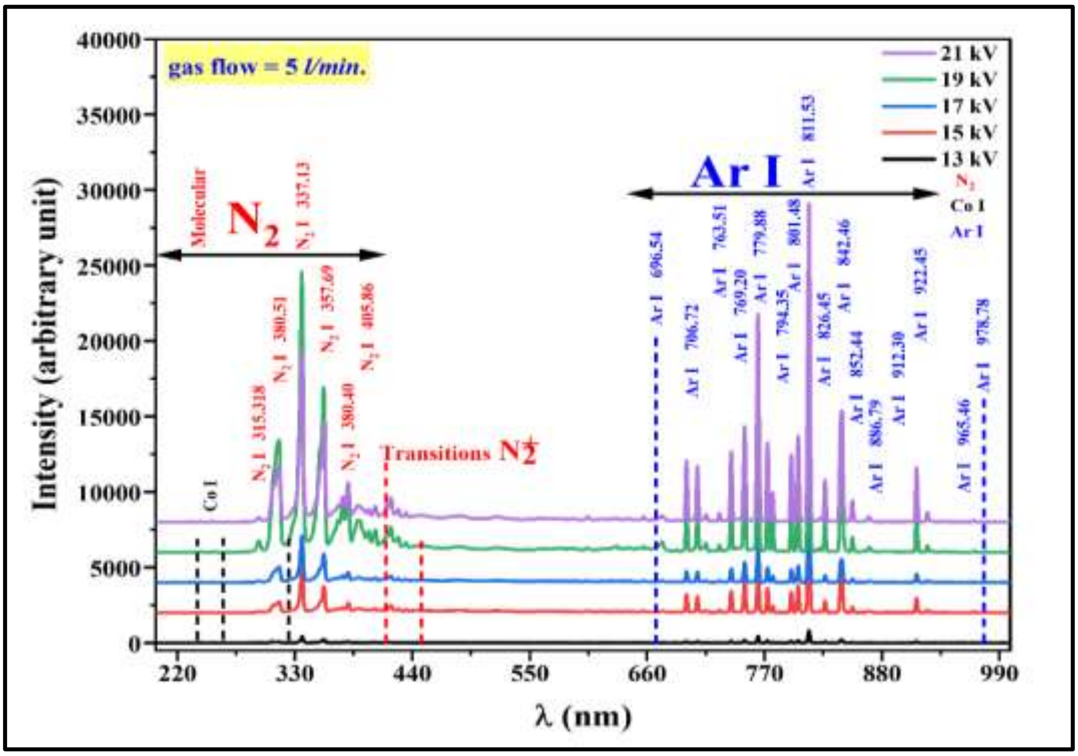

Figure 2: The distinction of intensity with wavelength for different values of applied voltage (13-21) kV.

Several peaks of argon gas are seen in the spectrum, most of which are in agreement with the data of the Higher Institute (NIST)[17]. The highest peak of the spectrum of the argon gas (ArI) appears at wavelength 811.5311 nm for all the values of the applied voltage; nitrogen gas (N_2) peaks appeared at the wavelength of (270-410) nm, with the highest peak at 337.13 nm wavelength at a flow gas rate of 5 *l/min*. The majority of the plasma peaks appeared in the (200-1000) nm wavelength range and there was an obvious increase in peak rates with the applied voltage at a constant argon gas flow rate of 5 *l/min.* These indicate an increase in plasma particle interactions and an increase in ArI gas emission[15]. The figure also shows the presence of active and generated plasma species when using this system's argon gas.

Figure 3 illustrates the peaks of cobalt metal (CoI) in the wavelength range of (230.901 – 255) nm, as they were diagnosed at different voltages (13-21) kV with the gas flow rate of 5 *l/min*. The results showed the appearance of the highest peak of (CoI), when using plasma, at a wavelength of 242 nm at the highest value of the applied voltage 21 kV compared and based on the data of the Higher Institute (NIST). The height of the peaks increased with increasing the applied voltages as shown in Figure 3.

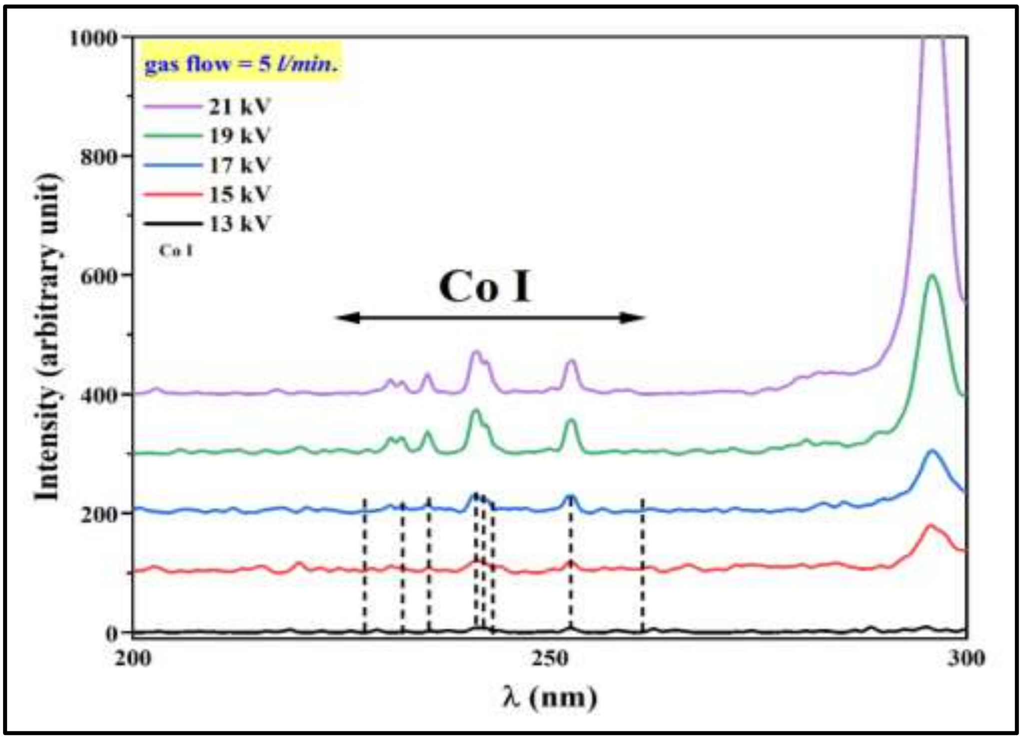

Figure 3: The intensity distinction for cobalt metal (CoI) as a function of wavelength for different values of applied voltage (13-21) kV.

The two figures below show the measurement of the number density and temperature of electrons, to determine the electron number density (n_e) which was calculated through Eq.4 as in Figure 4[18]. Through the data obtained for the highest peaks and when represented and plotted as $ln\left[\frac{\lambda_{ji}I_{ji}}{hcA_{ji}g_j}\right]$ with (E_j), the electron temperature (T_e) can be determined from the slope of the linear fitting of the resultant curve as shown in Figure 5 through using Eq.2.

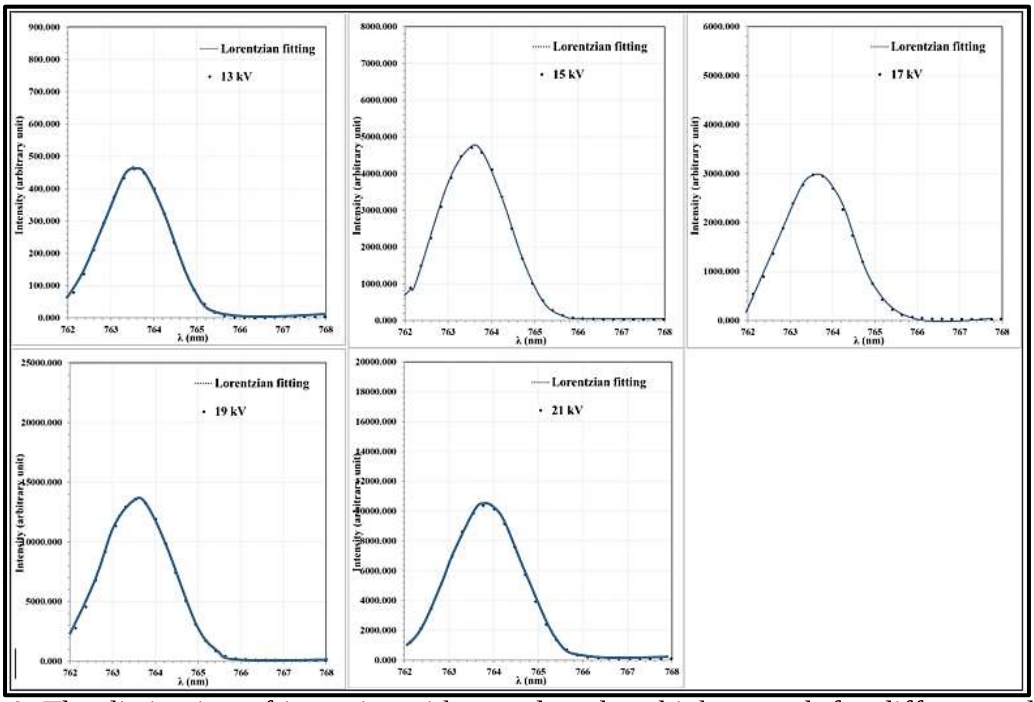

Figure 4: The distinction of intensity with wavelength at highest peak for different values of applied voltage.

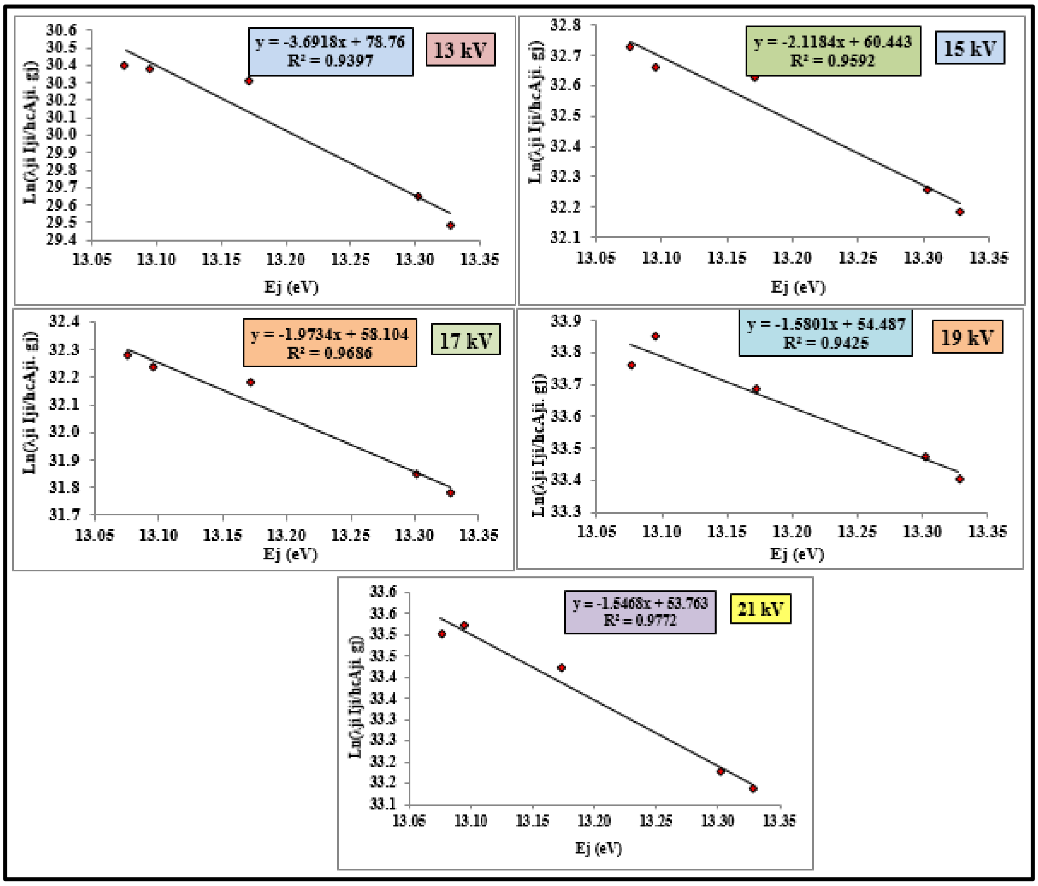

Figure 5: Boltzmann plots for different values of applied voltage (13-21) kV at the constant gas flow rate of 5 *l/min*

Figure 6 shows the agreement in the electron temperature as well as the electron density with the increase in the voltage applied or used in this system to generate plasma, where the results showed a clear increase in electron temperature from (0.27087 to 0.66498 eV) and electron density form (8.10811 x 10^{17} cm^{-3} to 13.85135 x 10^{17} cm^{-3}) with the increase of the applied voltage at a constant gas flow rate, as listed in Table 1.

Table 1: Atmospheric plasma jet parameters measured for different applied voltage (13-21) kV.

V (kV)	Te (eV)	FWHM (nm)	$n_{e*}10^{17}$ (cm^{-3})	f_p (Hz) *10^{12}	λ_D *10^{-6}(cm)
13	0.271	1.200	8.108	8.086	0.429
15	0.472	1.400	9.459	8.734	0.525
17	0.507	1.700	11.486	9.624	0.494
19	0.633	2.000	13.514	10.439	0.508
21	0.665	2.050	13.851	10.569	0.515

These results agree with those of Majeed et al.[19]. There are an increased number of collisions between electrons and argon atoms as the applied voltage is gradually increased, which results in an increase in the amount of energy transferred from electrons to gas molecules. This implies an increase in the number of argon gas molecules, which causes an increase in the number of collisions between the electrons and the gas atoms. As a result, the energy transferred from the electrons to the gas particles increases, causing an increase in the gas temperature. Also, the mechanism of excitation and ionization of atomic and ionic species in argon plasma is supposed to occur mainly by electron impact. So, when the argon gas flow rate increased, the high-energy tail of the electron energy distribution function was contracted to the lower energies. Therefore, the number of electron density increases when the flow rate increases due to the stepwise ionization inside the tube of plasma jet. In addition, this indicates that the energy supplied to the particles by the field is sufficient to cause secondary ionization of the molecules, thus, ionizing most of the argon gas molecules passing through the plasma tube. Therefore, the passing gas turns into a plasma state, consequently, the number of electron density appeared at the lowest value at the applied voltage 13 kV, whereas its highest value was at an applied voltage of 21 kV especially since argon gas flow is 5 *l/min*, and this indicates that the amount of argon gas passing through the plasma is too much.

It is clear that the measured plasma parameters are directly related to the applied voltage. As electrons are accelerated from the cathode to the anode with the kinetic energy of applied voltage they collide with the gas molecules present between the two electrodes, some of these electrons are absorbed by the neutral particles, while others are lost in the collisions (electron-neutral particle). As the applied field accelerates the newly liberated and colliding electrons, a secondary ionization process occurs, leading to the collapse process, which increases electron temperature and density[13], as shown in Figure 6. This leads to an electronic breakdown, resulting in an increase in voltage.

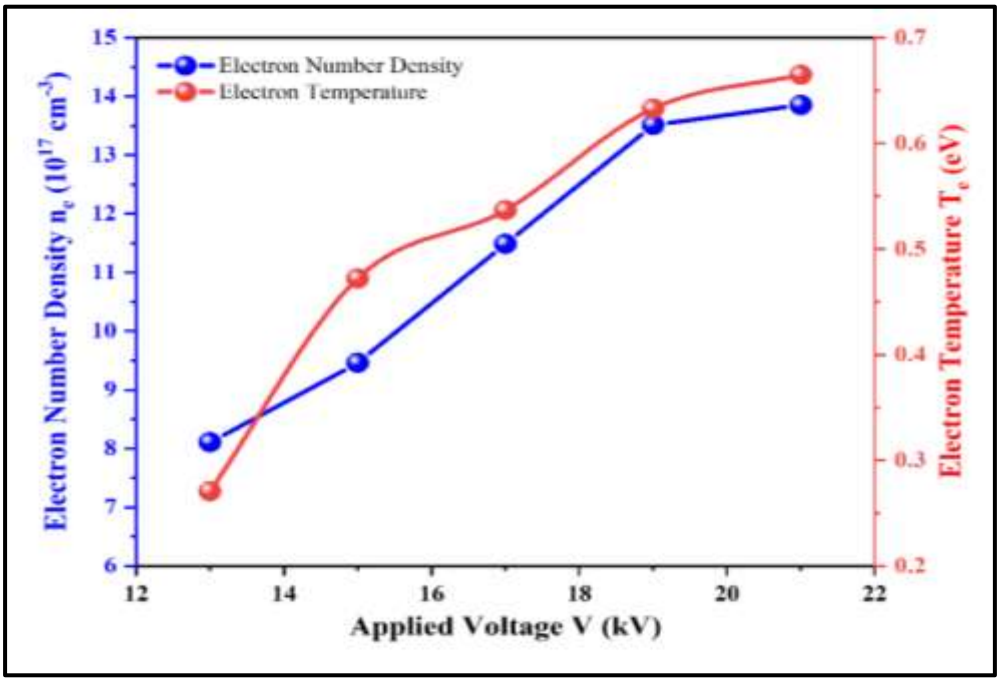

Figure 6: Relation of electron temperature and electron density with applied voltage (13-21) kV.

A comparison of plasma beam length generated in the atmosphere at various argon gas flow and at voltage levels of (13-21) kV can be seen in Figure 7 and 8. The plasma beam's maximum length of (3.65 cm 1.437 Inch) was obtained at the fifth flow for an applied voltage of 21 kV, while plasma plume lengths at the same flow for the other voltage levels of (13, 15, 17, 19, 21) kV were (2.1, 2.28, 2.75, 3.20, 3.65) cm, respectively[20]. In other words, as the applied voltage value increases, the amount of energy available to generate collisions between gas molecules flowing at each applied voltage value increases. Figure 7 depicts the relationship between the length of the plasma beam and the amount of gas flowing at each applied voltage value (13-21) kV. Thermal characteristics of the atmospheric plasma system are very important which can be used to develop this system for the preparation of nanotechnology for medical use or in medical aspect for accelerating the healing of wounds and burns. This agrees with Jiang and Bruggeman [21].

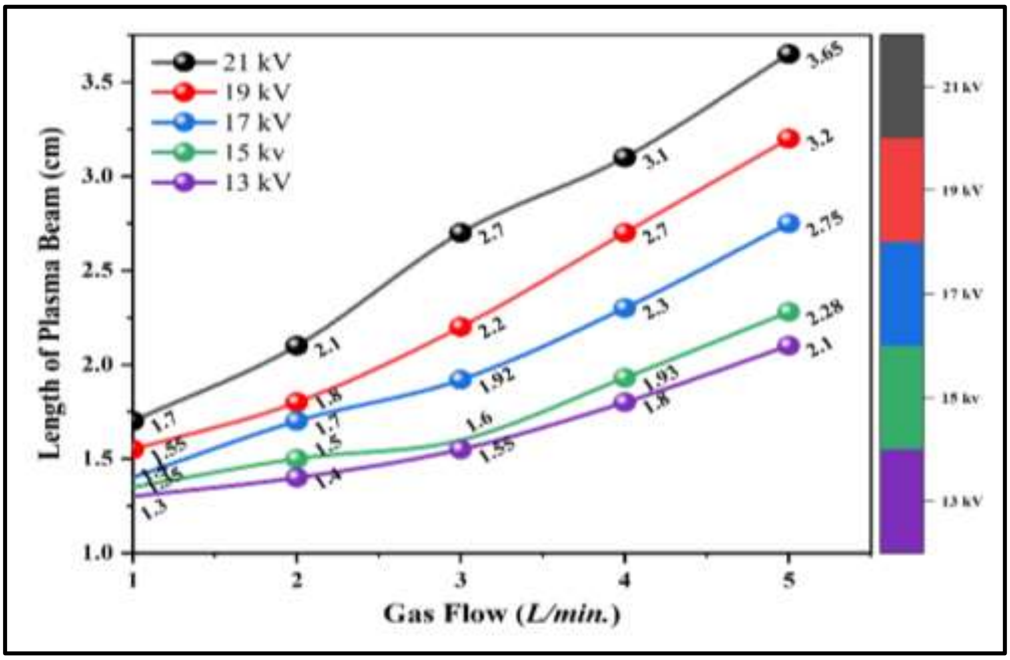

Figure 7: Length of the plasma beam as a function of the gas flow for different applied voltage (13-21) kV.

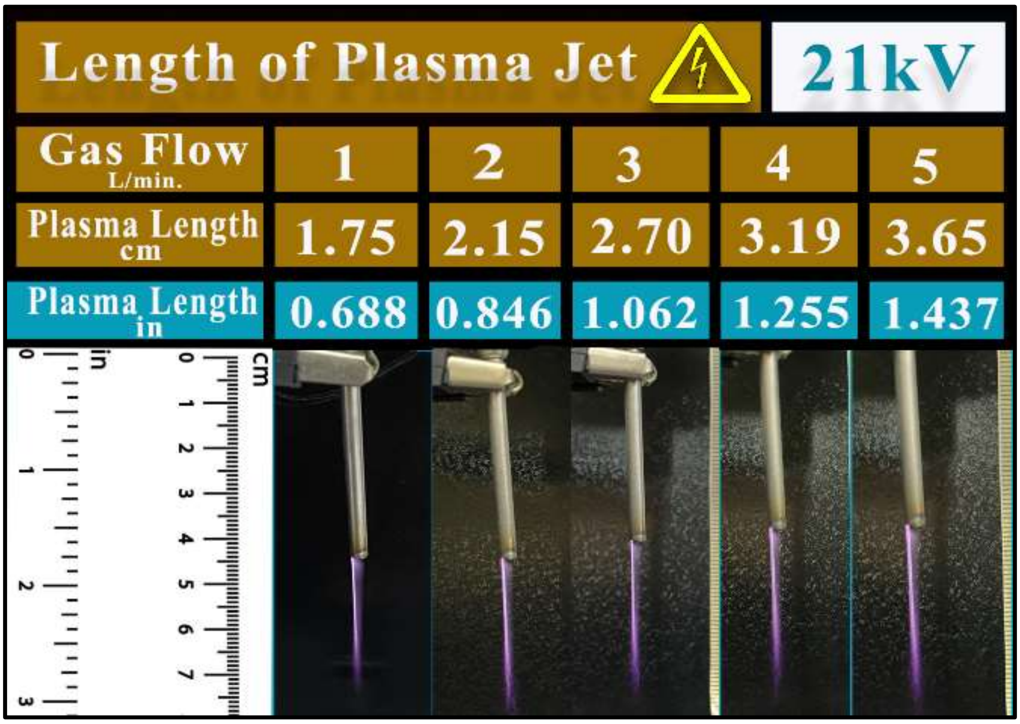

Figure 8: Cobalt plasma plume length of plasma jet at different gas flow (1-5) *l/min* of applied voltage 21 kV.

4. Conclusions

It was determined that the strength of the emission spectra lines of cobalt plasma created by a plasma jet is significantly dependent on external variables such as applied voltage and argon gas flow rate. The study found that increasing the applied voltage causes an increase in the intensity of emission, suggesting an increase in the number of gas molecules and number of collisions. Consequently, the effects of the applied voltage and gas flow rate on the excitation and ionization process on optical emission. Which means that the energy supplied to the particles is sufficient to cause secondary ionization of the molecules. The lowest intensity peaks of cobalt emission were at an applied voltage of 13 kV, whereas it is clear that its highest intensity peaks of cobalt emission were at an applied voltage of 21 kV, and this significantly affects the plasma parameters of the density and temperature of an electron, Debye length, and frequency of plasma which increases with the increase of the applied voltage. Furthermore, this system can successfully produce a length of plasma jet plume exceeding 3.5 cm at 5 *l/min* and 21 kV, which can be used in many applications.

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