Optimal Power of Atmospheric Pressure Plasma Jet with a Simple DBD Configuration for Biological Application

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Abstract: The new strategy, non-thermal atmospheric pressure plasma (N-APP), as a ‘physical’ method, could offer a simple, fast, effective, and economic way of disinfection of equipment, surfaces, a method that is unlikely to cause micro-organisms resistance, or allergic, and toxic reactions. Recently, cold physical plasma had been extensively studied by researchers as a possible therapy in dentistry and oncology, as well as the application of non-thermal plasma in biomedical researches such as wound healing. In clinical dermatology, cold plasmas are mainly used for the treatment of chronic wounds and pathogen-based skin diseases, in which stimulation of tissue repair and decontamination. In this research, the characterization of home-made Helium Non-Equilibrium atmospheric pressure plasma jet (He-NAPPJ), that had been generated using a dielectric barrier discharge (DBD) configuration for exceptional standardization protocol of this plasma source that meets medical requirements. The device equipped with two powered electrodes as well as a grounded electrode, driven by an (8 kVp−p) voltage, the frequency (12 kHz), and the distance between the nozzle of the plasma jet and the target were used as constant data. As a working gas, the Helium gas (He) was used in different flow rates (2, 4, 6, and 8) slm. The optical emission spectroscopy was used to measure the plasma parameters, of which the electronic excitation temperature and density of electrons were determined by the Boltzmann’s plot method and Stark broadening effect equation respectively. The result of the emission line spectrum showed the presence of nitrogen and oxygen between lines 300 nm until 700 nm. Nitrogen and oxygen are involved in the oxidation process which is known as Reactive Nitrogen species and reactive oxygen species. These species are the main key in bacteria inactivation and the wound healing process. Furthermore, the results had revealed, the optimal condition using the flow rate (6 slm) with 32 °C He gas temperature that had been stabilized for 20 min, plume length 40 mm, The distance is 15 mm from the distal end of the Pyrex tube (DBD) and the wound surface, the power density 44 mWatt/cm², and plasma radiation energy dose: 76 J/cm². These results were considered in this study as a safe operating condition for biomedical applications.

Keywords: Atmospheric pressure, cold plasma jet, electron temperature, the power density, Helium gas temperature, Plasma radiation dose.

1. Introduction:

Plasma, known as the fourth state of matter, plasma is a (partially) ionized gas in those ions, electrons, and photons are present as well as, radicals and molecules in an excited state [1]. The most commonly used method, of generating and sustaining low-temperature plasma, for technical
applications is by applying an electric field, to a neutral gas. Thermal and non-thermal (cold) plasmas, both already widely established in medicine, plasma applications hold big potential, for example in dermatology, to achieve the desired medical effect, especially in medical sterilization, and treatment of different kinds of skin diseases. Wound healing and tissue regeneration can be achieved by various types of plasma treatment, in a multitude of wound pathologies [2]. Atmospheric pressure plasma jets (APPJs), have indisputable advantages, such as environment-friendly substances, smaller generator, more economical and high stability, the ionization in non-thermal plasmas is not very high. But it is very effective in generating high concentrations of reactive radicals, of oxygen and nitrogen species (RONS) under room temperature conditions. Because of their rich reactive species, APPJs have become very attractive, for many applications in biomedical such as, bacterial inactivation, blood coagulation, cleansing of medical equipment, wound healing, and applications in dentistry [3]. The early experiments for the biological, and medical applications of plasma advanced a need for devices, that can deliver plasma outside the confinement of electrodes and enclosures arose. This need was met by the development of plasma sources, that can provide plumes of low-temperature plasma, outside the main discharge gap and into the ambient air. These devices are known as non-equilibrium atmospheric pressure plasma jets (N-APPJ), thus, various plasma jets were developed [4]. Non-thermal plasma (NTP) represents an effective tool, for various procedures in human as well as in veterinary medicine, particularly in tissue disinfection, and treatment of chronic wounds, such as diabetic foot ulcers, pressure, and venous leg ulcers, burns, and other skin pathologies, with microbial etiology [5,7]. Anti-itch, antimicrobial, anti-inflammatory, tissue-stimulating, blood flow-enhancing, and proapoptotic effects were demonstrated in vivo and in vitro experiments, and until now, no resistance of pathogens against plasma treatment was observed. Meanwhile, cancer treatments by APPJ. The non-thermal plasma is shown to be non-destructive to tissue, with only little effect on the surrounding (healthy) tissue [6]. In this research, we constructed a DBD cold plasma jet device operated at atmospheric pressure. The effect of electrical and optical characteristics of the plasma jet was investigated. The plasma dose and parameters of active particles presented inside the plume of plasma were analyzed by the OES method.

2. Experimental Apparatus and Methods:
2.1. Non-thermal Plasma Jet System (Experimental Setup):

In this study a home-made atmospheric pressure dielectric barrier discharge plasma jet Figure 1(A,B) device was used to generate non-equilibrium atmospheric pressure low temperature plasma.
2.2. Device Manufacturing:

The electrode system of the plasma jet consists of two parallel aluminum rings. The tubes were made of pyrex that represent a dielectric barrier discharge (DBD), the length and the inner diameter for tube was 95 mm and 1.9 mm, respectively, the thickness of the tube was 0.95 mm. The tube was covered by two aluminum rings electrode, where the thickness and width of the aluminum ring were 0.1 mm and 10 mm, respectively, this configuration was based on a double-ring electrode. The distance between two electrodes was fixed to 12 mm and the distance between downstream electrode and the nozzle of the pyrex tube was fixed to 2-4mm. The upstream electrode was connected to power supply and the downstream electrode was grounded [8], as shown in schematic diagram Figure 2.1 (b). Gas flow controller for Helium gas with commercial grade 99.999% (turkey) was fed via a flexible plastic tube, the flexible plastic tube was connected to the top edge of the pyrex tube by plastic lock while the flow rate of the gas was controlled by double stage regulator (BOC, UK) with flow meters 11420 (Mathesen, USA) and needle valve 288 01 B2 (Leybold, Germany).

2.3 Characterization of the Plasma Jet:
Measurement of the Plasma Plume Length:

The plasma plume length (the distance between the tip of the plasma plume and the edge of pyrex tube) or visible length of plasma plume into the ambient air was measured by a metric ruler, using the helium as working gas at (2, 4, 6, and 8) flow rates of operated gas, in applied voltage (8 KV) that used in this study.
Measurement of Plasma Plume Temperature:

The plasma jet temperature is one of the significant parameters which determine its applications scope. The temperature can be measured by using the mercury thermometer where the heat sensitive part was located in plasma plum at 15mm distances from the end of the tube nozzle for He gas at different gas flows.

Diagnostic of Plasma Plume:

One of the most commonly used methods to diagnose plasma is the optical emission spectroscopy (OES) technique. Plasma spectral diagnostic methods are based on measuring the intensity of the spectral lines of emission or absorption or the continuous spectrum, half widths and shifts of spectral lines, the emission spectra of the plasma jet was determined by ultraviolet visible-near-infrared (UV-Vis-NIR) spectrometer device SV2100 (Kmac, Korea) with wavelength range from 150 nm to 1000 nm. The spectrometer device was connected with an optical fiber cable M92L01 (Thorlabs, USA) to record the spectral emissions. To avoid the dispersion of plasma radiation, a collimator was used to assemble the radiation emitted from plasma. The collimator was located at 15 mm from the distal end of Pyrex tube, and it was connected to the optical fiber cable that linked to optical emission spectroscopy (OES) to record the spectral emissions. The data of spectral emissions was recorded by the spectrometer device connected to the laptop see Figure 1B[4, 5, and 13].

Measurement of the Electrical Discharge Parameters:

The instantaneous electrical powers $P(t)$ and total energy ($E$) dissipated in a pulse of He plasma discharge can be determined by[10]:

$$P = V I \cos \phi$$  \hspace{1cm} (1)

where $V$, $I$ are the RMS (root mean square) values of discharge voltage and current, respectively and $\phi$ is the phase shift between the measured voltage and the current signals (~ 0.95). The $V$–$I$ waveforms were recorded by a digital oscilloscope and the variations of the pulse-modulated condition had been studied for the correlation with the produced reactive species effects, because the application focuses on wound healing, gas temperature. To determine the mean power along the plume axis, the plasma plume voltage and current were measured at flow rate (2, 4, 6, and 8) slm using the voltage and the electric probe method, respectably. Figure (2) shows the $V$–$I$ characteristics of the developed He plasma jet: open-circuit voltage across the secondary coil of the transformer with 4 pulses in a burst (the voltage and current waveform of the discharge pulses). Calculation showed that the instantant electrical power (the mean power at the substrate) was 17W. On other hand, the average power dissipated was determined using the voltage ($v(t)$) and current ($i(t)$) from the grounded copper tape in this experiment[10].
\[ P_{Diss} = \frac{1}{T} \int_0^T v(t)i(t)dt, \] ............ (2)

where T is the period of applied voltage (70 µsec.). From measured current and voltage, calculations showed that the dissipated power was 1.4 mW.

Measurement of treatment plasma dose:

To estimate plasma radiation in terms of an optical power (W/cm²) for continuous exposure of the skin generated by the He plasma jet, the spectral region in the 252nm UV range of the He plasma jet was measured using the bench top optical power/energy meter (Newport Inc. model 842.PE Canada.) with photodiode sensor (200–1100 nm). The photodiode sensor was located at 5 mm from the nozzle tip of the developed He plasma jet, as shown in Figure (3).

3. The Results:

3.1. Electrical Characteristics:

AC sinusoidal voltage with a peak-to-peak value Vp–p of 8 kV at a fixed frequency of 12 kHz is applied to the power electrodes and the helium gas (gas flow rates of 2, 4, 6 and 8 slm) was generated through the quartz tube, the discharge is ignited and spurted into the ambient air abruptly. Directly at the end of the funnel-shaped nozzle, a bright channel follows, which pale sheath surrounds. The plasma plume length was studied as a function of gas flow rate with helium gas at different flow rate and exposure times in the case. The length of plasma plume measured by metric ruler. To describe the development of plasma plume length, the maximum length of Helium gas, the plasma plume reached (44 mm) at (4 slm), in addition the width of the plasma plume in helium increased when higher gas flow rate and high voltage were used. The results revealed the flow (6 slm) of helium gas, that was produced a constant non-thermal plasma plume in plume length (40 mm).
3.2. Plasma Plume Temperature:

Figure 4 demonstrated that plasma jets temperature depends on working gas flow rate. The room temperature was 28°C during all the measurements. It was observed that the plasma jet plume temperature raised with rising time and plasma jet temperature decrease with increased gas flow rate, that can be attributed to the effect of thermal conductivity of the gas. Further, the gas temperature at all flow rates reached thermal equilibrium after 2min of plasma exposure and remained the same up to 20 min. The thermal conductivity can affect highest temperature, it was (0.15) W/(m K) for helium gas. The results revealed the flow (6 slm) of helium gas, that was produced a constant non-thermal plasma plume in low gas temperature 32 °C for 20 min.

![Figure (4): shows the statistical relationship between helium gas temperatures at different gas flows](image)

3.3 Plasma Parameters:

The active species of plasma jet and the diagnostics of plasma parameters ($T_e$ and $n_e$) were exhibited by using OES method. Figure 5 (a,b) shows the spectral lines of helium plasma jet which emitted in the UV-Vis regions. It is observed that the emissions of atomic helium with peaks at 388.9 nm and 706.5 nm have stronger intensity, while emissions with peaks at 447.1 nm, 501.4 nm, 587.5 nm, 667.8 nm and 728.1 nm are relatively weak. A variety of active species such as OH, N$_2$, N$^+$, NO, O II, H and O I are clearly observed in emission spectrum of the helium plasma jet.
Electron Temperature and Density:

Boltzmann plot method was employed in this work to determine the $T_e$ for Helium plasma jet. Electron density was determined from the Saha-Boltzmann equation after knowledge of the $T_e$ by using atomic and ionic spectral line [10].

$$n_e = 6.04 \times 10^{21} \frac{\Omega_{\text{He}}}{\Omega_{\text{He}} \gamma_{	ext{He}}} A^J \times \exp \frac{E^J - E^{\text{corr}} - E^J}{k_B T_e}$$

Figure (6) illustrate the Boltzmann plot using 3 lines of He I, to increase the accuracy, for different gas flow rates. The selection lines are isolated and presence in all curves. The $T_e$ were calculated using equation from Boltzmann plot method from the inverse of slope of best liner fitting for the relation between

$$\text{Ln} \left( \frac{I_{\Omega_{\text{He}}}}{h c \gamma_{\text{He}} \Omega_{\text{He}}} \right) \text{versus upper energy level (Ej)} \quad (4)$$

The equations of fitting were shown in the figure (6). Figure (7) Shows the 706.5nm He I, peak profile where full width at half maximum(W) found by using Lorentzian fitting to calculate electron density ($n_e$) at different gas flow rates using Stark broadening effect equation [11]. The values compared with standard values in the national institute of standards and technology (NIST) atomic spectral database:

Where $W=0.0074$nm and $n_e = 1.00E+16$ (cm$^{-3}$)
Depending on the standard values of broadening for this line [12], it can be seen that the full width increase with increasing gas flow rates from 2 to 4 slm and then start to decrease at flow rate 6 and 8 slm which indicate on increasing electron density and then starts to decrease with the increased gas flow rates.

![Boltzmann plot](image)

Figure (6): The Boltzmann plot using 3 lines of He I, to increase the accuracy, for different gas flow rates

![Peak profile](image)

Figure (7): The 706.5nm He I, peak profile where full width at half maximum found by using Lorentzian fitting to calculate electron density at different gas flow rates
The variation of electron temperature ($T_e$), electron density ($n_e$) and plasma parameters with gas flow rates were shown in Figure (8) and table (1).

![Figure (8): The relationship between electron temperature ($T_e$), electron density ($n_e$) and Helium gas flow rates.](image)

| Flow rate (l/min) | $T_e$ (eV) | FWHM (nm) | $n_e \times 10^{17}$ (cm$^{-3}$) | $f_p$ (Hz) $\times 10^{12}$ | $\lambda_0 \times 10^{6}$ (cm) |
|------------------|----------|-----------|-------------------------------|----------------------------|-------------------------------|
| 2                | 0.321    | 4.400     | 4.400                         | 5.957                      | 0.634                         |
| 4                | 0.267    | 4.600     | 4.600                         | 6.091                      | 0.566                         |
| 6                | 0.253    | 4.700     | 4.700                         | 6.156                      | 0.545                         |
| 8                | 0.237    | 4.800     | 4.800                         | 6.222                      | 0.522                         |

The plasma dose:

The estimation of plasma radiation in terms of an optical power (W/cm$^2$) generated by the He plasma jet, was measured using the bench top optical power/energy meter. The mean power density for 1x1cm$^2$, and energy dose for the treatment sample area is constant (3.14 mm$^2$) along the plume axis, were illustrated in table (3.2). Based on the experimental matrix, a dose of 1 minute at 44mWatt/cm$^2$ for each square cm was deemed maximum acceptable prolonged treatment.
Table (2): The radiation dose produced by helium gas plasma jet at different gas flows.

| He gas Flow (l/min) | Power density (mWatt/cm²) | Energy dose: (J/cm²) |
|---------------------|---------------------------|---------------------|
| 2                   | 350                       | 660                 |
| 4                   | 50                        | 95                  |
| 6                   | 44                        | 76                  |
| 8                   | 20                        | 38                  |

4. Discussion:

To obtain the optimal conditions for biomedical applications of the plasma jet, the characterization of the homemade DBD-APPJ device was made. Generally, standardization of devices is in the focus of the physical plasma to compare and identify key process parameters, plasma components, and experimental conditions. The first step in success by translating plasma source protocols and biological application to reach a unique standardization protocol, which was, based on the characterization of the plasma jet. The plume length and the temperature for and helium plasma jets were measured. The effect of applied voltage and gas flow rate on the plasma parameters (Te and ne) for helium plasma jet was determined. In addition, the dependence of line intensity of active species on gas flow rate and applied voltage were investigated. In a study by [13], that mentioned, the effect of peak-to-peak voltage increase can be explained in terms of the higher local electric field produced in the plasma jet. The higher electric field produces a higher electron concentration which, in turn, enhances the production of active species and therefore increases the emission intensities. Meanwhile, increasing the gas flow rate leads to increased intensity of spectral lines, which means a higher amount of reactive plasma species are present and that is because of the increase in gas flow rate result in a lower mole fraction of air which surrounds the plasma plume. The lower mole fraction of air can lead to higher local electron concentration, which in turn increases the emission intensities of the active species [13].

In this study one type of electrode configuration was used, the device equipped with two powered electrodes. Pyrex tube was used as DBD configuration, the inner diameter was (2mm) and the pyrex tube wall thickness was (0.95 mm), which was used as the best thickness for the pyrex tube of this home-made DBD-APPJ device, that was reported in the previous study by Hammoodi [8], who used the same device, and found that the plasma plume length showed a clear increase when reducing the thickness of the tube wall, furthermore, the plasma jet temperature had decreased when reducing the thickness of the tube wall. Characterization of home-made DBD APPJ device was made by setting the
power supply at the system conditions: a high-voltage (8 kVp−p) and high-frequency (12 kHz), the operating gas used was helium gas, the gas was used in different flow rates (2, 4, 6, and 8) slm, and the plasma plume temperature was measured for each gas flow, at (0, 30 sec., 2 min., 6 min, 10 min, 14 min, 18 min, and 20 min), to study the stability of plasma plume temperature during 20 min., also, to reach the low-temperature plasma radiation suitable for wound healing. The distance used was (15 mm) between the plasma jet exit and the measurement tools (thermometer or OES). Ghimire et al.,[14] Suggested that the decrease in the distance leads to the generation of more active species which play an important role in the inactivation process of bacteria. On the other hand, reducing the distance between the plasma jet exit and the target increases the temperature. Therefore, the distance from the plasma jet exit should be optimized to not exceed the temperature above 43⁰C which can lead to cell damage.

The results illustrated that the plume length in helium and the width of the plasma plume in helium increased when higher gas flow rate and high voltage. Moreover, there were two regions of plasma plume were observed in Helium plasma: a bright straight region in front of the Pyrex tube and an irregular region at the tip of the jet. The first zone is observed at gas flow rates from 2 slm to 6 slm, which corresponds to the laminar flow. The second zone is observed at 8 slm, which corresponds to the turbulent flow. The turbulent flow caused instabilities and deformations in the plasma plume [15]. As a result, the length of the plasma plume decreased by 6 slm for helium plasma jets, the same finding was obtained by [9] when used (7.5 kVp−p) as the working voltage was found the length of the plasma plume decreased by 5 slm for helium plasma jet.

Moreover, the result in this study showed a constant non-thermal plasma plume was produced in low He gas in flow rate (6slm) that can reach the biologically tolerable temperatures, (T < 40 °C) is better than other flow rates used. Paschen’s law can be utilized here to explain the gas pressure which is related to flow rate and the influence of voltage on the length of the plasma column. The particle concentration in the plasma tube is relatively lesser at a lower flow rate (gas pressure), which leads to a lower collision rate between the electrons and molecules. At a high flow rate, the mean free path of the electrons is too short to gain enough energy to ionize the gas molecules. Hence, the flow rate and pressure have the most appropriate value for an electron avalanche. According to the law, the statistic breakdown voltage (Us) is higher in both the low and high flow rate and pressure cases. So as the flow rate increases, the breakdown voltage decreases at first, and then rises. The overvoltage U−Us = U rises at first and then decreases at a fixed applied voltage, which results in that the length of the plasma plume first increases and decreases afterward as the flow rate increases. Similarly, setting the flow rate at a specific value, increasing only the voltage leads to a larger U, which results in the growth of the plasma plume. In addition to that, a larger U means the electrons get more energy initially [16].

The results were observed in Figure (5 a, b) that the higher applied voltage and gas flow rate results in the richness of these active species lines. As well as, it appears that the active species lines
are affected by an increased gas flow rate more than the applied voltage. In addition, the effect of peak-to-peak voltage increase can be explained in terms of the higher local electric field produced in the plasma jet. The higher electric field produces a higher electron concentration which, in turn, enhances the production of active species and therefore increases the emission intensities in the homemade helium plasma jet. The same results have been found in a study of Al rawaf [9], which used the same device for bacterial inactivation. Meanwhile, increasing the gas flow rate leads to increased intensity of spectral lines, which means a higher amount of reactive plasma species are present. This is because the increase in gas flow rate results in a lower mole fraction of air that surrounds the plasma plume. The lower mole fraction of air can lead to higher local electron concentration, which in turn increases the emission intensities of the active species [13]. The results showed that the Te for helium plasma jets decreases slightly with increasing gas flow rate from 2 to 8 slm. This behavior can be attributed to the Reynolds number. This number increase with increasing gas flow rate. The higher Reynolds number leads to increase gas density, which in turn reduces the mean free path. The reduction of the mean free path can lead to a decrease Te because of increased electron-neutral collision frequency. Furthermore, the dependence of Te and ne for helium plasma jets on the applied voltage. This behavior can be attributed to the effect of the electric field, where the higher voltage leads to an increase in the intensity of the electric field between the aluminum electrodes, and hence electrons collect more energy from the electric field. As a result, Te increases with the increasing applied voltage [17].

The UV radiation is a useful sterilization mechanism, especially in the 200-300 nm wavelength range where the UV radiation in this range is mostly associated with DNA damage [19]. In helium plasma jets, most of the UV radiation will be reabsorbed in the atmospheric pressure and will not reach the sample surface, which is justified in Figures (5 a, b), where there is no evident UV emission spectrum between 200-300 nm. It indicates that UV radiation is not responsible for the process of bacteria inactivation. The helium plasma jet is a rich source of RONS. As shown in Figure (5 a, b), the line intensity of OH radical shows evidently in the helium plasma jets. The OH radicals have a direct effect both on the membrane and cell wall of bacteria. In addition, the reactive oxygen species (ROS) such as atomic oxygen cause an oxidative stress response leading to lipid peroxidation, inactivation of mononuclear proteins, and DNA damage, which eventually cause harmful oxidative cell damage [20]. Lin et al [21], reported that the OH radicals are much more reactive than other ROS
from the aspect of redox biology and have a shorter diffusion distance, which makes OH radicals significant in decontamination bacteria.

As the plasma dose is a very important parameter in biomedical applications, there are critical values that should be taken into account to avoid cell damage. Dobrynin et al. [22] postulated that plasma dose < 135 J/cm² is a safe dose for biomedical application. The plasma dose depends on both sample exposure time and power density i.e.; the plasma dose is the product of these two factors [23]. According to Wu et al [23], the higher levels of the power density (0.31 W/cm²) can cause damage in the living tissue at low exposure times (1 min), meanwhile, lower power density with long exposure time (0.13 W/cm² for 15 min) does not affect the living tissue. Therefore, it is important to estimate the power density to study the safe operating conditions of the plasma source to be applicable for biomedical applications. It is observed in the table (2) that the power and related energy of plasma dose decreases with increasing gas flow rate. The plasma plume temperature and related energy dose depend on the gas flow characteristic. The plasma plume temperature is observed to decrease as the gas flow rate is increased. This seems to be mainly attributed to a rise in collision frequency of hot plasma species with the surrounding cold molecules, effectively leading to a reduction in the plume temperature [24]. These results indicate that the plume temperature and plasma energy dose can be adjusted effectively by controlling the gas flow rate. A recent study by Lou [25], was in the same line with this study who evaluated the effect of a non-thermal plasma jet in the treatment of cutaneous wounded male Sprague-Dawley (SD) rats. The working voltage was 7.5 kV, sample distance 20 mm, and working gas Helium and argon gases in treatments, respectively. Various RONS generated by non-thermal plasma sources. Based on the optical emission spectroscopy (OES) spectra of He-based cold atmospheric pressure plasma jet (CAPJ), the compositions and abundances of RONS generated by CAPJ are most likely dependent on the composition of the working gas.

**Conclusion:**

The results represent a non-thermal atmospheric pressure plasma jet in DBD configuration as an effective tool in the development of plasma sources that can provide plumes of low-temperature plasma outside the main discharge gap and into the ambient air, to achieve biologically tolerable temperatures, (T < 40 °C) and this plasma jets was conceder as small and low-cost plasma sources, also NAPPJ can overcome the limitations of the size of materials or living tissues like skin wounds to be treated. Moreover, it provides an acceptable plasma radiation energy dose for bio-medical applications.

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