Determination of Helium-Discharge Atmospheric-Pressure Plasma Parameters and Distribution Using Numerical Simulation

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This study investigated the chemical distribution of an atmospheric-pressure plasma jet (APPJ) along its propagation direction using numerical simulation. Low-resolution spectral data were used to estimate the gas temperature and the excitation temperature. These estimations were used with a collisional-radiative model to elucidate population densities and the electron temperature. A global model was applied to investigate the chemical species distribution in the plasma jet. The thermodynamic properties of the APPJ corresponded well to the relation $T_g < T_{\text{exc}} < T_e$ for all the positions along the jet propagation. Chemical species generation and propagation along the plasma jet were numerically simulated using the GM with input parameters derived from the CR model and the ideal gas law.

Keywords: atmospheric-pressure plasma, numerical modeling, chemical species distribution, spectral characterization

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1. Introduction

Atmospheric-pressure plasma (APP) has attracted considerable attention owing to its applicability in a wide range of environmental, chemical, biomedical, and material processes [1–5]. The non-thermal properties of APP are suitable for temperature-sensitive applications. Chemical species generation through APP also increases its potential use in applications such as surface treatment, sterilization, and nanoparticle synthesis. Generation of APP jets (APPJs) using dielectric or metal capillaries has been widely studied in the biomedical and material-processing fields because of its feasibility in plasma patterning for surface treatments and focusing APP to a local position [6–10]. Although APPJ properties are important to their applications, their qualitative properties are difficult to study from experimental observations. Therefore, numerical modeling is used to predict the properties of APP [11–15].

The behavior of APPJs is important for understanding thermodynamic properties including gas temperature ($T_g$), rotational temperature ($T_{\text{rot}}$), excitation temperature ($T_{\text{exc}}$) and electron temperature ($T_e$) in the plasma [16–19]. Optical emission spectroscopy (OES) is a widely used technique to investigate such properties using experimental observations [20–23]. The accuracy of estimating such thermodynamic properties varies with the resolution of the obtained OES [24–27]. APPs are known to be derived from non-local thermodynamic equilibrium conditions to non-equilibrium between the rotational temperature $T_{\text{Rot}}$, the excitation temperature $T_{\text{exc}}$, and the electron temperature $T_e$. The rotational temperature $T_{\text{Rot}}$ provides information about the gas temperature $T_g$. The use of OH lines or the second positive systems of N$_2$ (C–B, 0–0, or 0–2) are widely used techniques to estimate $T_g$ using OES information [28–30]. Furthermore, the use of intensity of working gas or stark broadening can be used to estimate $T_{\text{exc}}$ and $T_e$, respectively [31,32]. However, estimation of these thermodynamic properties using OES requires a high-resolution spectrometer. Other than the use of high-resolution spectrometers, $T_g$ and $T_{\text{exc}}$, which are estimated from the excitation level population, can be used with a collisional radiative (CR) model to estimate $T_e$ [33–36]. The distribution of the chemical species along APPJs provides vital information to improve their applicability. Experimental investigations of these chemical distributions can be performed using the localized OES of the plasma jets. However, the determination of such properties is a challenging process. Therefore, numerous studies have used numerical modeling to predict the possible chemical distributions along APPJs [12, 15, 37–39]. A global model (GM) is one of the modeling techniques widely used to estimate the time evolution of chemical species of APPJs [37–40].

In this study, we focused on investigating the ther-
modynamic behaviors and chemical species distributions along APPJs using a CR model and GM. Optical emission lines collected from a double-electrode configured APPJ were used with a general approach on $N_2$ (C–B, 0–0) lines to estimate $T_g$. The excitation temperature ($T_{exc}$) describes the population of atomic excited states assuming that this follows a Boltzmann distribution. Considering the degeneracy of each atomic state, the population come by [18, 19, 41], and the CR model was applied to estimate the relations between the thermodynamic properties of APPJs. The distribution of chemical species along the direction of plasma jet propagation was estimated numerically using the GM.

2. Numerical Modeling Methodology

2.1 CR model with atomic collisions

Optical emission spectra provide information about the atomic process in excited atoms. These experimental observations with numerical simulations can provide details about the plasma parameters. To analyze such processes, the population of excited levels in the working gas of APP must be estimated. For such work, CR models, which were first introduced by Bates et al. [42, 43], and Johnson et al. [44], have been extensively used.

In CR models, rate equations are written for the modeled energy levels of atoms or ions, and various processes that affect the excited and de-excited electron number density of the modeled levels are simulated. Therefore, in CR models the population number densities of the ground state and modeled excited states are calculated along with the free electron density [22, 35, 45–47]. However, to generate the APPJs inter-atomic collision is important rather than the free electron collision. In this study, the noble gas helium was introduced into the equipment. The collected localized optical emission lines were used to estimate the propagation of $T_g$ and $T_{exc}$ with the plasma jet. Estimated values were input into the CR model to calculate the $T_g$ and population number densities of the helium gas.

As an APPJ is considered to be a weakly ionized plasma, the population number densities of the excited level $N_p$ (where $p \geq 2; p = 1$ corresponds to the ground state) were approximately estimated using the CR model as shown in Eq. (1):

$$\frac{dN_p}{dt} = C_{1p}N_eN_1 + N_1 \sum_{j=1,j\neq p}^{M} K_{jp}N_j - N_pN_1 \sum_{j=1,j\neq p}^{M} K_{pj}$$

$$= 0, \quad (p \geq 2) \quad (1)$$

where $M$ is the upper limit of the excitation level included in the CR model. The matrix and vectors are defined as follows:

$$\mathbf{K} = \begin{pmatrix}
    K_{2j} & -K_{32} & \cdots & -K_{M2} \\
    -K_{23} & K_{3j} & \cdots & -K_{M3} \\
    \vdots & \vdots & \ddots & \vdots \\
    -K_{2M} & -K_{3M} & \cdots & \sum_{j=1}^{M} K_{Mj}
\end{pmatrix}, \quad (2)$$

$$N = \begin{pmatrix}
    N_2 \\
    \vdots \\
    N_M
\end{pmatrix}, \quad (3)$$

$$C_1 = \begin{pmatrix}
    C_{12} \\
    \vdots \\
    C_{1M}
\end{pmatrix}, \quad (4)$$

$$K_1 = \begin{pmatrix}
    K_{12} \\
    \vdots \\
    K_{1M}
\end{pmatrix}, \quad (5)$$

For the excited-state number density $N_p = (p = 2, 3, 4, \cdots , M)$, we rewrite Eq. 1 as

$$\mathbf{KN} = N_eC_1 + N_1K_1.$$  

(6)

We know that the electron temperature of the plasma and gas temperature are considered in Kelvin, and the electron temperature is much higher than the gas temperature. For most problems in plasma, the atomic collision excitation from the ground state is basically negligible. Hence, $N_eC_1 \gg N_1K_1$, and Eq. 6 can be approximated to

$$\mathbf{KN} = N_eC_1.$$  

(8)

However, we must confirm the steadiness of the matrix $\mathbf{K}$ in Eq. 2. The collisional de-excitation from the level $p$ to the ground state $K_{pi}$ in the ground state in the summation of diagonal elements is negligibly small in general. If the diagonal element does not include $K_{pi}$, the matrix $\mathbf{K}$ becomes always singular for any matrix elements. Then the summation of every row vector in the matrix becomes exactly a zero vector. Fortunately, owing to the addition of the term $K_{pl}$ into the summation of each diagonal element in Eq. 2, $\mathbf{K}$ generally becomes a regular matrix. Furthermore, for low-gas-temperature conditions, the atomic collisional de-excitation to the ground state $K_{pl}$ ($j \neq 1$) cannot be neglected. Thus, we always have an algebraic solution of the vector $N$ from Eq. 8. Hence,

$$N = N_e\mathbf{K}^{-1}C_1,$$  

(9)

where the matrix $\mathbf{K}^{-1}$ depends only on $T_e$ and the vector $C_1$ only on $T_0$. The gas temperature has almost been studied by the rotational temperature in APPJs [28, 29]. Even in pure helium plasma, we can detect molecular band spectra
such as a second positive system of $N_2$ molecule transition. In the next step, we fixed the gas temperature and used the relation of the excitation temperature from the experimental result to determine the electron temperature using Eq. 10:

$$N_p = \sum_{i=1}^{M} \frac{[K^{-1} (T_g)]_{pi}}{[K^{-1} (T_g)]_{qi} C_{1i}(T_e)} = F(T_g, T_e).$$

We found the value of the gas temperature and subsequently could derive the electron temperature by spectroscopic analysis of the excitation temperature together with the CR model calculation. From the CR model, we estimated the population number density of helium in this study by using the atomic data of helium provided in Table 1 from the atomic spectra database of the National Institute of Standards and Technology [48].

### 2.2 Global model

To comprehend the chemical reactive species in APPs, zero-dimensional plasma chemical kinetics simulations such as GMs are widely used [49], as the chemical species generated through APPJs play a major role in improving the applicability of such techniques. This study also considered the use of GM to estimate the behaviors of the chemical species along the APPJs. The obtained results were compared with experimentally observed results.

For this work, we considered 25 main species, as listed in Table 2, and 56 possible reactions were adopted from the work by Murakami et al. [39] for the numerical simulation. Our numerical simulation was carried out in time dependent model. It assumed that the plasma moves by the constant drift velocity determined by the introduced gas flow. The governing rate equation considered in this study is expressed as follows:

$$\frac{d n_i}{d t} = n_i \sum_{j=1}^{25} k_{ij} n_j + n_i \sum_{j=1}^{25} n_j n_{m} k_{ijm} + R_{ex},$$

where $n_i$, $n_j$, $k_{ij}$, $k_{ijm}$ and $R_{ex}$ denote time, number density of $i$th species, two-body reaction rate for $i$ and $j$th species, three-body reaction rate for $i$, $j$, and $m$th species and external rate source term, respectively [14, 39, 50].

Even though the GM predicts the time evolution of chemical species, this study considered the position evolution of chemical species to understand the distribution of such species along a plasma jet. To convert the time evolution to position evolution, we considered the flow behavior of the APPJ in the laminar regime and estimated the laminar flow velocity to estimate the propagation time and position, as follows:

$$\frac{1}{\tau} = \frac{v_f}{d}.$$  

Where $\tau$ is the time considered in the time evolution estimation in the GM, $v_f$ is the laminar flow velocity of the plasma jet and which is 42.26 m/s is given by the experimentally obtained from the gas flow rate considering with the cross-section of the tube [10, 51], and $d$ is the position corresponding to the relevant time evolution. We assumed that the flow velocity of the plasma was constant in the calculation.

### 3. Results and Discussion

#### 3.1 Thermodynamic parameter estimation using low-resolution spectroscopy

OES provides important information on the thermodynamic properties of APPJs. The gas and excitation temperatures are the thermodynamic properties widely explained using experimentally obtained OES. In this study, we also estimated the above thermodynamic properties using experimentally obtained OES. The spectra were obtained in the range of 200 - 800 nm using a spectrometer system (HAMMAMATSU: PMA-12) with a spectral resolution $\Delta \lambda < 2$ nm.
Fig. 1 Determination of gas temperature from synthetic spectra for the N$_2$ (C–B, 0–0) transition.

The gas temperature $T_g$ can be estimated directly using high-resolution OES from the rotational temperature $T_{\text{Rot}}$. Although low-resolution OES was mainly used to detect the possible radicals and emission lines of the plasma, this study estimated $T_g$ using averaging of summing of theoretical intensities over the experimentally detected lines from the equilibrium between the gas temperature $T_g$ and the rotational temperature $T_{\text{Rot}}$.

The nitrogen second positive system N$_2$ (C$^2\Pi_u \rightarrow$ B$^3\Pi_g$, 0–0) 337 nm line is the commonly used emission line system to estimate the rotational temperature or $T_g$ of the plasma state [28, 29, 52]. In this work, N$_2$ (C–B, 0–0) simulated values were summed and averaged over the spectrum around $\lambda \pm (\Delta \lambda)/2$ of the experimentally obtained OES lines. The relative intensities of these values were then considered and fitted by a least-squares method along the experimentally obtained OES spectrum for an absolute $T_g$ value. The gas temperature was changed and proceeded further until the simulated shape closely coincided with the experimental spectral curve, as shown in Fig. 1. The line broadening of the simulated fitting curve expands with the increase in gas temperature. The $T_g$ value that most closely synthesized the experimental spectrum was considered the rotational temperature of the plasma state at that position.

This method was applied to estimate $T_g$ along the direction of plasma jet propagation, and the distribution of $T_g$ along the jet position is shown in Fig. 2. As shown in Fig. 2, $T_g$ was approximately 400 K for the helium APPJ at the nozzle exit as possible expected values [53]. In addition, as the plasma jet propagated, $T_g$ was also observed to decrease toward room temperature at the bottom of the plasma jet. In addition to $T_g$, the He excitation temperature can also be estimated using the low-resolution emission spectra. In this study, to determine $T_{\text{exc}}$, a Boltzmann plot was drawn with the quantity $\ln(I_{ij}/\lambda_{ij} g_i)$ against the energy of the corresponding energy of the He lines, where $i$ and $j$ are the upper and lower levels, respectively. Recorded correspondence intensities ($I_{ij}$) and wavelength ($\lambda_{ij}$) were used with the Einstein coefficient ($A_{ij}$) and statistical weight ($g_i$) obtained from the National Institute of Standards and Technology (NIST) [48]. He spectra of 389, 471, 587, 667, 706, and 728 nm were used to calculate $T_{\text{exc}}$, and the information related to each line is listed in Table 3.

Using the localized OES data with the Boltzmann plot method, $T_{\text{exc}}$ was estimated along the direction of plasma jet propagation, as shown in Fig. 3. $T_{\text{exc}}$ is high at the nozzle exit and tends to decrease as the jet propagates. Such behavior along the plasma jet can be expected as the relaxation of rotational levels and metastable excited levels of helium decrease.

Table 3 Excitation energy and transition strength of helium.

| Wavelength (nm) | Configuration [48] | Energy (eV) [48] | Einstein A
|-----------------|--------------------|-----------------|-------------
| 389             | 1s3p $^3P$        | 1s2s $^3S$      | 23.0711     | 9.47E-06 | 106 ns |
| 471             | 1s3p $^3P$        | 1s2p $^3P$      | 23.5939     | 3.17E-06 | 315 ns |
| 587             | 1s3d $^3D$        | 1s2p $^3P$      | 23.0737     | 4.20E+07 | 23 ns  |
| 667             | 1s3d $^3D$        | 1s2p $^3P$      | 23.0741     | 2.12E+07 | 47 ns  |
| 706             | 1s3s $^1S$        | 1s2p $^3P$      | 22.7185     | 9.28E-06 | 102 ns |
| 728             | 1s3s $^3S$        | 1s2p $^3P$      | 22.9203     | 1.83E+07 | 54 ns  |

Fig. 2 Gas temperature estimated by synthetic spectrums for the N$_2$ (C–B, 0–0) transition along the plasma jet.

Fig. 3 Excitation temperature distribution along the plasma jet.
3.2 Estimation of population number density, electron temperature, and electron density

Electron density and electron temperature are important parameters that describe the plasma state. Akatsuka et al. [22, 35] explained the probability of population density in argon plasma with the presence of excited-level populations and excitation kinetics of nonequilibrium ionizing argon discharge plasma at atmospheric pressure [22, 35]. Estimating the parameters of $n_e$ and $T_e$ using the low-resolution emission line spectra is a difficult process. Therefore, in this study, as we were able to estimate $T_g$ and $T_{exc}$ using the experimentally obtained OES data, the CR model was adopted to estimate the $T_e$ and population densities of the working gas. The estimated $T_{exc}$ along the plasma jet in this study fell in the range 0.51 - 0.57 eV and, according to the CR model, these values relate to low electron temperature values. Analysis of the relationship between the lower electron temperature and excitation was carried out in this work. For this purpose, we consider the relation between the thermodynamic properties $T_g$, $T_{exc}$, and $T_e$ in the CR model, as shown in Fig. 4.

The electron temperature for certain $T_{exc}$ and $T_g$ was estimated using this method to understand its propagation along the plasma jet. Figure 5 shows the propagation of $T_{exc}$ and $T_e$ along the plasma jet for the corresponding $T_g$ at each position.

Although the electron temperature decreases along the direction of jet propagation, the relation between the gas temperature, excitation temperature, and electron temperature maintains the condition of $T_g < T_{exc} < T_e$ along the direction of jet propagation. This information confirms that the considered plasma jet maintains its non-equilibrium condition along the plasma jet.

Many studies have estimated the electron densities of helium plasma in APPJs using the stark broadening method [3, 4]. As our OES system has low resolution, this study used the CR model to numerically estimate the population number density for helium excited states. In addition, this study used the relation between the electron density and population density state in Eq. (9) to estimate the electron number density for this simulation.

According to the CR model estimations, the relation between $T_e$ and $n_e$ shows a similar shape for different electron densities against electron temperature, as shown in Fig. 6. However, the curves shift to higher values as the electron density increases. Using the information from the helium excited energy level states in Table 1, the population number density of each level was numerically estimated using the CR model against $T_e$, as shown in Fig. 7. For this numerical estimation, experimentally obtained $T_e$ and $T_g$ values for the plasma jet at the nozzle exit were used with the CR model at an electron density of $10^{11}$ cm$^{-3}$. Ex-

![Fig. 4](image1.png)

Fig. 4 Calculation of excitation temperature as a function of electron temperature from the CR model with level pairs at $n_e = 10$ cm$^{-3}$, $T_g = 340$ - 400 K at $P = 1$ atm as a function of electron temperature.

![Fig. 5](image2.png)

Fig. 5 CR model calculation of the distribution of excitation and electron temperatures along the plasma jet.

![Fig. 6](image3.png)

Fig. 6 Shows the relation between the He$^+$ density as a function of the different from $10^9$ to $10^{15}$ cm$^{-3}$ electron density with $T_g = 400$ K.
Experimental evidence of electrons density for helium APPJs tends to be around $10^{11}$ cm$^{-3}$ [39]. According to the model calculation, the triplet states of the helium energy levels show higher population densities compared to the singlet states. Therefore, the emission lines between triplet states should have high intensity compared to the singlet transitions. Experimental work conducted by Fernando et al. [10, 51], parallel to this study, confirmed that triplet transitions have the highest emission intensities for He plasma. Using the estimated thermodynamic profiles along the plasma jet with the CR model, this study numerically estimated the population number density distribution, as shown in Fig. 8. Figure 8 clearly shows a decrease in the excited level population number densities as the plasma jet propagates. As per our experimental observations, we also noted that the intensity of the helium emission lines decreased along the plasma jet [10, 51]. This can be identified as helium excited atoms tending to relax as the APPJ propagates. As per the CR model, the population density strongly depends on the electron temperature.

### 3.3 Estimation of chemical distribution along the APPJ

We used a GM to numerically simulate the distribution of chemical species along the jet propagation. The GM described by Murakami et al. [39] was considered and numerically simulated using the initial and estimated parameters. The population density of excited level $2p^3P$, which was estimated for an electron density of $10^{11}$ cm$^{-3}$, was used as the initial condition of excited helium density as per the CR model estimation and observed emission lines. The time evolution estimation was converted to the position evolution using Eq. (11) to describe the behavior of the chemical species distribution along the plasma jet. The estimation of chemical species was also focused mainly on the above-mentioned radical estimation. For this purpose, the initial input parameters of electron density and densities of the chemical species of helium, nitrogen, oxygen, and H$_2$O ppm were estimated considering the atmospheric conditions of the experiment. The chemical species distribution along the plasma jet was numerically simulated using the aforementioned parameters using the GM. The distribution of species along the plasma jet is shown in Figs. 9 - 11.

As illustrated in Figs. 9 - 11, the chemical species exhibit sudden generation around the nozzle exit before propagating along the plasma jet. The interaction of air with the helium plasma plume at the nozzle exit and nanosecond-scale reaction rate are the main reasons for the sudden increase in species at the nozzle exit.
Figure 12 illustrates the normalized distribution of chemical species around the nozzle exit. As the densities of chemical species are of different orders, the highest individual density value for the defined time scale was used to normalize the distribution of chemical species.

Although this study uses an electron density of $10^{11}$ cm$^{-3}$ as the initial condition, further investigation was performed to understand the behavior of chemical species in relation to electron density is shown in Figs. 13, 14. Figures 13 and 14 illustrate the behavior normalized He$^+$ and OH distribution as a function of the difference from $10^{11}$ to $10^{15}$ cm$^{-3}$ electron density along with the plasma jet. The He$^+$ and OH density could be significant for using comparison with experiment work. This study will be the focus of our future study.

4. Conclusions
In this study, we investigated the behavior of APPJs through numerical modeling. Low-resolution spectroscopy data were used to estimate the gas temperature and the excitation temperature was estimated along the jet propagation. As per the numerical estimation, the gas temperature was approximately 400 K at the nozzle exit and
The thermodynamic properties of the APPJ were experimentally obtained gas temperature and excitation temperature in relation to the position of the plasma jet with the experimental data [10]. The estimation of the excitation temperature showed a higher value at the nozzle exit with a tendency to reduce in value as the APPJ propagated. The CR model for the helium working gas was used to estimate the electron temperature and population densities derived from the CR model and the ideal gas law.

The CR model for the helium working gas was used to estimate the electron temperature and population densities in relation to the position of the plasma jet with the experimentally obtained gas temperature and excitation temperature data [10]. The thermodynamic properties of the APPJ corresponded well to the relation $T_g < T_{exc} < T_e$ for all the positions along the jet propagation. Chemical species generation and propagation along the plasma jet were numerically simulated using the GM with input parameters derived from the CR model and the ideal gas law.

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