Determination of the electron temperature and density of non-equilibrium atmospheric-pressure Ar plasma by OES using two-tempareture EEDF

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Abstract: A two-temperature electron energy distribution function (EEDF) is proposed for the non-equilibrium argon atmospheric-pressure plasma. Its continuum emission spectrum, dominated by electron-neutral bremsstrahlung radiation, is analyzed to determine the electron temperature (T_e) and density (N_e) by optical emission spectroscopic (OES) measurement. T_e was 0.4 eV and N_e were 3.3×10^{15} cm⁻³ with a discharge voltage of 5.4 kV. The two-temperature EEDF provided a better fit than the one-temperature EEDF.

Keywords: optical emission spectroscopy, two-temperature electron energy distribution function.

1. Introduction

Novel non-equilibrium atmospheric-pressure plasma (NEAPP) applications are discovered rapidly across a wide variety of fields. For instance, NEAPP have been utilized in the fields of materials engineering [1], and medicine [2]. NEAPP can be applied to various materials because it is operated at low gas temperatures and does not require a vacuum equipment. However, the electron temperature (T_e) , electron density (N_e) , and electron energy distribution function (EEDF) have not been reported frequently. Therefore, its simulation cannot be performed to estimate densities of reactive species. Hence, it is necessary to determine these parameters.

The plasma analysis method in this paper is Optical Emission Spectroscopy (OES). OES does not disturb the plasma unlike probe measurements. By measuring the continuum spectrum due to electron-atom bremsstrahlung, to determine the T_e and N_e in the NEAPP is possible [3 -4]. However, to determine the T_e and N_e , the EEDF is required. Since the plasma is in a non-equilibrium state, the EEDF does not follow the Maxwellian EEDF. Previous studies have concluded that determining the EEDF is difficult in such cases. A good EEDF has been reported in two types. First, one-temperature Druvesteynian EEDF was better than one-temperature Maxwellian EEDF [5]. However, Fitting is difficult in the long-wavelength region. Second, by using machine learning, a two-temperature Maxwellian EEDF provides good results [6]. However, this method has the drawback of poor wavelength resolution. Based on these considerations, EEDF can be considered as a two-temperature EEDF of Maxwellian and Druyvesteynian.

2. Theoretical analysis

At atmospheric pressure, the continuum spectrum is clearly measurable by OES. The continuum spectrum mainly consists of recombination radiation and bremsstrahlung for the NEAPP. Under atmospheric pressure, the ionization degree of the plasma can be assumed to be below 10^{-3} . Therefore, electron-neutral bremsstrahlung is the dominant source of the continuum spectrum [3 - 4]. The emissivity of electron-neutral bremsstrahlung can be expressed below [4].

$$\epsilon_{\rm ea} = \sqrt{\frac{2}{m_{\rm e}} \frac{N_{\rm e} N_{\rm a}}{\lambda^2} \frac{hc}{4\pi} \int_{\frac{hc}{\lambda}}^{\infty} Q_{\rm ea}^{\rm B}(\lambda, E) \sqrt{E} F(E) dE} \qquad (1)$$

Here, ϵ_{ea} is the electron-neutral bremsstrahlung emissivity, F(E) is the EEDF, Q_{ea}^{B} is the electron-neutral bremsstrahlung cross section, and $E, m_e, N_e, N_a, h, \lambda$, and c are the electron-energy, electron mass, electron number density, neutral particle number density, Planck constant, wavelength, and speed of light, respectively.

One-temperature EEDF is assumed to be formulated as follows [7 - 8]

$$F(E) = \gamma \frac{\Gamma\left(\frac{5}{2\gamma}\right)^{\frac{3}{2}} \sqrt{E}}{\Gamma\left(\frac{3}{2\gamma}\right)^{\frac{5}{2}} \left(\frac{3}{2}kT_{e}\right)^{\frac{3}{2}}} \exp\left[-\left(\frac{\Gamma\left(\frac{5}{2\gamma}\right)E}{\Gamma\left(\frac{3}{2\gamma}\right)\frac{3}{2}kT_{e}}\right)^{\gamma}\right] (2)$$

In the case of gamma equal to 1, F(E) is Maxwellian EEDF, while for gamma equal to 2, it is Druyvesteynian EEDF. Two-temperature EEDF is calculated as a linear combination of one-temperature EEDF. Thus, two-temperature EEDF is the following equation.

$$F_{\rm mix}(E) = \alpha_1 F_1(E) + \alpha_2 F_2(E)$$
 (3)

Here, F_{mix} is the two-temperature EEDF, whilst F_1 , and F_2 is the one-temperature EEDF. α_1 , and α_2 is the Mixing ratio. F_1 , and F_2 have their temperature of T_{e1} , and T_{e2} , respectively. T_{e1} and T_{e2} is electron temperature, and can be different from each other. When fitting the experimental data, the T_e and N_e can be obtained as variables by adjusting T_{e1} , T_{e2} , α_1 , and α_2 as parameters. An EEPF is obtained by dividing EEDF by \sqrt{E} . In the graph of EEPF, Maxwellian EEPF is given as a linear function and Druyvesteynian EEPF is given as a quadratic function.

3. Experiment

Figure 1 shows a schematic overview of the dielectric barrier discharge NEAPP generator in this experiment. Further details are described elsewhere [5].

The Ar gas flow rate is adjusted with a flow controller and set to 4 L/min to ensure the stability of the discharge. Discharge voltage is applied with a high-voltage power supply as an inverter-type neon transformer, up to 9.0 kVpp as AC 20 kHz. In the present study, the output voltage V was set as 4.5 - 8.1 kV in 0.9 kV steps. A spectrometer (MS3504, SOL Instruments Ltd., Czerny- Turner mount, focal length 350 mm, F/3.8, used grating 1200 grooves/mm, blaze wavelength 500 nm) is connected to the optical fiber guide tube. The wavelength sensitivity was calibrated in advance using a standard irradiance light source and a white diffuse reflection element for converting it to a radiance standard. However, this measurement was performed without long-pass filter, and consequently, it may be mixed by higher-order diffraction light.



Fig. 1. Schematic diagram of this experimental setup [5].

4. Results and discussion

Figure 2 shows the fitting result of emissivity with a discharge voltage of 5.4 kV. The best theoretical value is 0.37 eV Druyvesteynian and 0.90 eV Maxwellian mixed at a ratio of 9999:1. The average electron temperature is 0.37 eV. It is dominated by Druyvesteynian. Electron density is found to be 3.3×10^{15} cm⁻³. Figure 3 shows the EEPF used in the theoretical calculation. In the low electron-energy range, the EEPF is predominantly Druyvesteynian, while in the high electron-energy range, it is Maxwellian. The fitting becomes improved compared to previous studies owing to its successful fitting over a wide range of wavelengths [5]. Based on these results, it is revealed that this plasma contains a high number of high-energy electrons despite their low electron temperature.

Figure 4 shows the variation of electron temperature and electron density as the discharge voltage is changed. The averaged electron temperature did not change. Concerning the Maxwellian part, which corresponds to high-energy tail, the temperature decreased with the voltage. The electron density increased with increasing the voltage. This result is similar to the previous studies [6]. One possible explanation for the decrease in the temperature in highenergy tail with an increase in voltage is as follows. The number of collisions between electrons increased, because of the increase in its electron density. Therefore, the energy relaxation among electrons has progressed, resulting in a decrease in the number of high-energy electrons.



kV.



Fig. 3. The EEPF used in the theoretical calculation in Figure 2.



Fig. 4. Best-fitted results at various applied voltages.

5. References

- [1] T. Yuji, and Y.-M. Sung, *IEEE Trans. Plasma Sci.* **35**, 4, 1010-1013 (2007).
- [2] T. Yuji, K. Nakabayashi, H. Kinoshita, N. Mungkung, Y. Suzaki, S. Mamat and H. Akatsuka, *Journal of Food Quality* **2021**, 8896716 (2021).
- [3] S. Park, W. Choe, S. Y. Moon, and J. Park, *Appl. Phys. Lett.* **104**, 084103 (2014).
- [4] K. T. A. L. Burm, *Plasma Sources Sci. and Technol.* 13, 387 (2014)
- [5] H. Onishi, F. Yamazaki, Y. Hakozaki, M. Takemura, A. Nezu, and H. Akatsuka, *Jpn. J. Appl. Phys.* **60**, 026002 (2021).
- [6] T. van der Gaag, A. Nezu and H. Akatsuka, *Jpn. J. Appl. Phys.* **61**, 076004 (2022).
- [7] J. T. Gudmundsson, *Plasma Sources Sci. Technol.* **10** 76 (2001).
- [8] J. B. Boffard, R. O. Jung, C. C. Lin, and A. E. Wendt, *Plasma Sources Sci. Technol.* **19**, 065001 (2010).