Current Applied Physics 10 (2010) 1456–1460

Contents lists available at ScienceDirect

Current Applied Physics

journal homepage: www.elsevier.com/locate/cap

Parametric study on excitation temperature and electron temperature in low pressure plasmas

Hoyong Park, Wonho Choe*

Department of Physics, Korea Advanced Institute of Science and Technology, 335 Gwahangno, Yuseong-gu, Daejeon 305-701, Republic of Korea

ARTICLE INFO

Article history:
Received 12 May 2010
Accepted 25 May 2010
Available online 31 May 2010

Keywords:
Excitation temperature
Electron temperature
Optical emission spectroscopy
Langmuir probe

ABSTRACT

This work aims at investigation of the validity of the electron excitation temperature (T_{exc}) by optical emission spectroscopy (OES) as an alternative diagnostic to the electron temperature (T_e). The excitation and the electron temperatures were measured at a wide range of gas pressures and input powers in different plasmas such as capacitively-coupled, inductively-coupled, and magnetron direct current plasmas. As a result, both temperatures were found to decrease with an increase in pressure, whereas they not very dependent on power, indicating that T_{exc} showed a tendency identical to that of T_e as pressure and power were varied. This result suggests that T_{exc} measurement can be an alternative diagnostic for T_e measurement once the ratio of the two temperatures is found in advance through a calibration experiment especially for low pressure high electron density industrial processing plasmas in which probe measurements are limited.

1. Introduction

Electron temperature (T_e) is one of the most informative and basic parameters in plasma physics because electrons are not only involved in the excitation, dissociation, and ionization of atoms and molecules but also govern the chemical reactions inside the plasma. The kinetic temperature of the free electrons is usually related to the electronic excitation temperature (T_{exc}) of the bound electrons in an atom or a molecule because excitation processes governing the distribution of excited states are mostly caused by the free electrons [1]. Excitation temperature is widely utilized in high pressure plasmas, and, especially, T_e determination at an atmospheric pressure is frequently based on the measurement of T_{exc} due to their close relationship [1–3].

The electron temperature of the plasma is measured by a variety of diagnostic techniques, and one of the most prevalent and versatile means of measuring T_e is the electrostatic Langmuir probe [4]. Despite its simple hardware configuration, its utilization can be sometimes limited due to the difficulty in analysis especially in the presence of negative ions, a possible origin of plasma perturbation, and its difficult use for large volume plasmas. Optical emission spectroscopy (OES) can also determine T_e based on appropriate equilibrium models [5–7]. However, it is sometimes not straightforward to obtain information corresponding to collision cross-sections of all species and to determine T_e directly from the spectral lines due to cumbersome interpretation using complex equilibrium models.

On the other hand, T_{exc} measurement, which is directly evaluated from atomic emission lines, is sometimes more efficient to characterize the plasma in low pressure plasmas as well as in high pressure plasmas. The reasons for this are that this characteristic temperature is relatively easy to determine and that it equals T_e in the case of equilibrium. When the electron density is higher than the critical one given by the Griem criterion, the system obeys the local thermodynamic equilibrium (LTE) and T_{exc} is then close to T_e [8]. Although it would be difficult to expect $T_e = T_{exc}$ in laboratory or industrial processing plasmas because they seldom satisfy the LTE condition, elucidating the relationship between the two physical parameters will be very instructive, especially for plasmas in which direct T_e measurement is difficult.

The relationship between T_{exc} and T_e has been investigated over the past few decades, and the resulting studies reported the comparisons performed in microwave discharge [2,3,9], plasma jet [10], or theoretically [11] at atmospheric pressure. However, few studies have been attempted in high electron density at low pressure plasmas such as industrial processing plasmas in which various plasma sources are needed for advanced technology.

In this paper, we investigated the relationship between T_{exc} and T_e in a wide range of electron density (from 5.2×10^9 to 3.2×10^{11} cm^{-3}) in different low pressure plasmas sources including capacitively-coupled plasma (CCP), inductively-coupled plasma (ICP), and magnetized direct current plasma (MDCP) sources. As a result, we found that T_{exc} has the same tendency as T_e when the gas pressure and input power were varied.

* Corresponding author.
E-mail address: wchoe@kaist.ac.kr (W. Choe).

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doi:10.1016/j.cap.2010.05.013

Section 2 describes the experimental setup, and the results and discussions for both the excitation and electron temperatures are presented in Sec. 3. This is followed by the conclusion in Sec. 4.

2. Experiments

Measurements were performed with a typical OES setup and Langmuir probe, as illustrated in Fig. 1. The plasma sources employed in the experiments were CCP, ICP, and MDCP sources, respectively. In the CCP source [12], the plasma was generated between two parallel electrodes 300 mm in length with a gap distance of 40 mm in a rectangular vacuum chamber. The upper electrode was powered through an impedance matching network from 50 W to 500 W at a fixed frequency of 13.56 MHz using a radio frequency (rf) generator (YOUNGSIN, XSR-06MF), while the lower electrode was grounded. The argon operating pressure ranged from 27 mTorr to 600 mTorr. In the ICP source [13], a flat coil antenna 20 mm in diameter was positioned on a quartz window 30 cm in diameter and 2 cm in thickness. The antenna was powered through an impedance matching network from 200 W to 800 W at 13.56 MHz using an rf generator (ENI, A1000-1971). The argon operating pressure was varied from 1 mTorr to 30 mTorr. In the MDCP source [14], a magnetron sputtering system for deposition of titanium film was used in the experiment. Argon gas was used to generate the plasma and the titanium emission from the circular titanium target with a diameter of 50 mm, which acts as the cathode of the glow discharge, was used to obtain T_{exc}. The target was powered from 150 W to 300 W with dc voltage and current monitored while the substrate was floating. Magnetic field was measured to be about 300 G at the target by a Hall probe Gaussmeter. The operating pressure was varied from 20 mTorr to 50 mTorr.

The optical emission from the plasma was measured by a CHROMEX 250 is spectrometer with a 250 mm focal length and 1200 groove/mm, which gave a spectral resolution of 0.375 nm with an entrance slit width of 50 μm. The spectrometer was scanned from 400 nm to 810 nm with a charge-coupled device (CCD) detector (SBIG ST-9XE). The collimation optics with two convex lenses was calibrated using a mercury lamp. As depicted in Fig. 1, two convex lenses 500 mm in focal length were used to focus the same diagnostic area of the probe measurement on the entrance slit of the spectrometer. The electron temperature was obtained from the I–V characteristic curve by using the rf compensated Langmuir probe (two peak impedances at 13.56 MHz and at 27.12 MHz) in both CCP and ICP, respectively, and by a single Langmuir probe in MDCP.

3. Results and discussions

For excitation temperature T_{exc} measurement, the Boltzmann plot method was employed. The atomic emission intensity (I_{jk}) of the transition from level j to level k depends on the Einstein coefficient of spontaneous emission between the excited levels of j and k (A_{jk}) and absolute population of the atomic level (n_j), as follows:

$$I_{jk} = n_j A_{jk} h\nu, \quad (1)$$

where h is the Plank constant and ν is the photon frequency corresponding to the transition from level j to level k. Assuming a Boltzmann distribution of the population of the atomic levels, I_{jk} is given by [2]

$$I_{jk} = \frac{hc}{\lambda}\frac{n g_j A_{jk}}{U(T_{exc})}\exp\left(-\frac{E_j}{T_{exc}}\right), \quad (2)$$

which is rearranged as

$$\ln\left(\frac{I_{jk}\lambda}{A_{jk}g_j}\right) = 1.4388 \times \frac{E_j[\mathrm{cm}^{-1}]}{T_{exc}[K]} + \mathrm{const.}, \quad (3)$$

where n is the number density of bound electrons in all ionization states, $U(T_{exc})$ is the partition function, c is the speed of light in free space, λ is the wavelength of the corresponding transition. Also E_j and g_j are the energy difference and the statistical weight of a level j, respectively. The Boltzmann plot is based on Eq. (3) to represent the natural logarithm of I_{jk} against the energy E_j, in which each data point corresponds to each emission line of a specific wavelength λ. The excitation temperature T_{exc} is evaluated from the inverse of the slope of the Boltzmann plot.

For T_e measurement using the Langmuir probe, the electron energy probability function (EEPF), $g_p(\varepsilon)$ was experimentally obtained from the second derivative (d^2I_e/dV^2) of the probe current–voltage characteristics as follows:

$$g_p(\varepsilon) = g_e(\varepsilon)\varepsilon^{-1/2} \propto d^2I_e/dV^2, \quad (4)$$

where $\varepsilon = -eV$ is the electron energy and $g_e(\varepsilon)$ is the electron energy distribution function (EEDF). In this experiment, the electron energy was measured as the mean electron energy $\langle\varepsilon\rangle$ by integrating the measured EEPF, and the electron density n_e was also measured by the same method.

As expressed in Eq. (4), EEPF is obtained from the EEDF and can be expressed against the electron energy and electron temperature for a Maxwellian distribution,

$$g_p(\varepsilon) = \frac{2}{\sqrt{\pi}} n_e T_e^{-3/2} e^{-\varepsilon/T_e}, \quad (5)$$

which is rearranged by taking the natural logarithm of EEPF against the electron energy as below [15] so that we get,

$$\ln g_p(\varepsilon) = \mathrm{const} - \frac{\varepsilon}{T_e}. \quad (6)$$

The electron temperature was found from the inverse of the slope of Eq. (6), which is the same procedure for obtaining T_{exc} in Eq. (3).

Fig. 2 shows the results of T_{exc} by OES (solid curves) and T_e by probe (dotted curves) measurements as a function of gas pressure from 25 mTorr to 600 mTorr and input power from 100 W to 500 W in CCP. In addition, the ratios between T_{exc} and T_e as a function of the gas pressure and input power are shown, respectively.

The decrease in T_e with increasing pressure at a fixed power is typical and has been reported elsewhere [16]. However, there is a steep increase in both T_e and T_{exc} at low pressure from 27 mTorr to 40 mTorr in Fig. 2(a). This can be explained by abnormal heating in terms of the bounce resonant motion of low energy electrons in the bulk plasma in the low pressure capacitively-coupled plasmas [17].

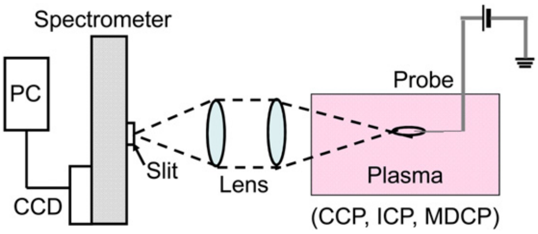

Fig. 1. A schematic of the experiment setup consisting of a Langmuir probe and an optical emission spectroscopic arrangement. Two convex lenses were used to focus the plasma image on the probe tip location to the entrance slit of the spectrometer.

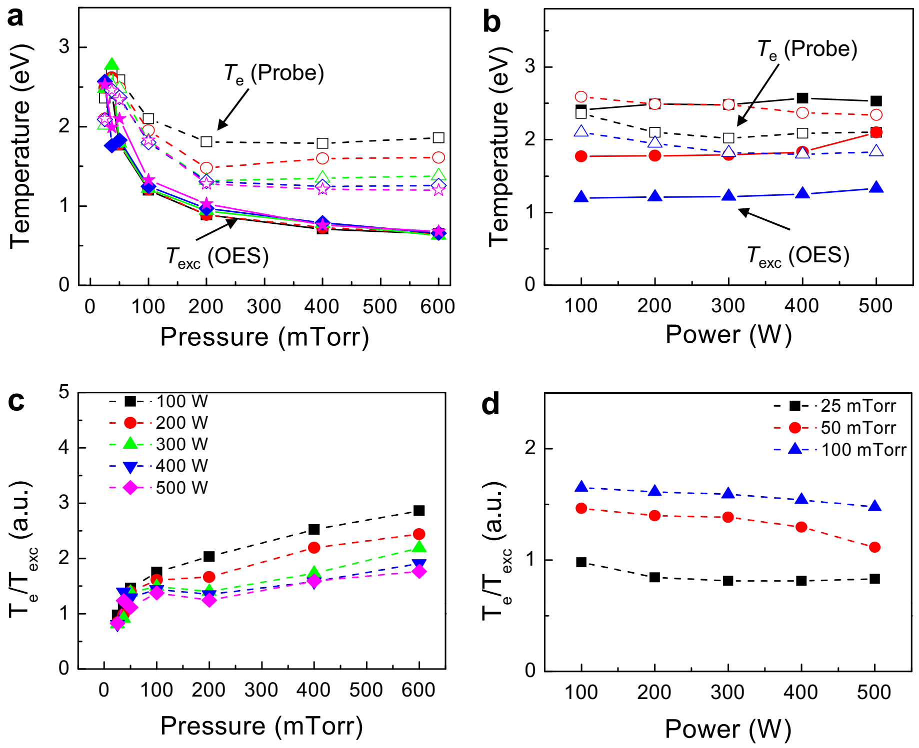

Fig. 2. The measured results of T_{exc} by OES (solid curves) and T_e by probe (dotted curves) as a function of (a) gas pressure (25 mTorr–600 mTorr) and (b) input power (100 W–500 W) in CCP. The ratios T_e/T_{exc} as a function of (c) gas pressure and (d) input power, respectively, are given.

Note also that the power dependence of both T_{exc} and T_e is weaker than the gas pressure dependence as shown in Fig. 2(a) and (b). This is because T_e is determined by particle conservation and is independent of the plasma density and, therefore, the input power [4,15]. The temperature ratios T_e/T_{exc} depicted in Fig. 2(c) and (d) suggest that T_{exc} measurement can be an alternative diagnostic for T_e measurement once the ratio of the two temperatures is found in advance through a calibration experiment.

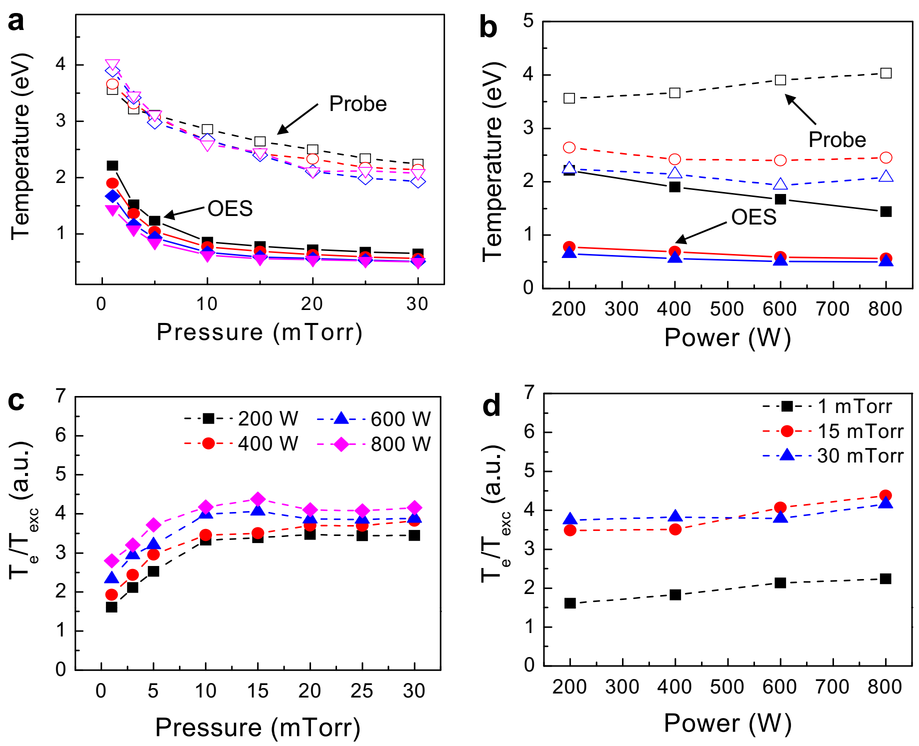

Fig. 3. Similar results for ICP.

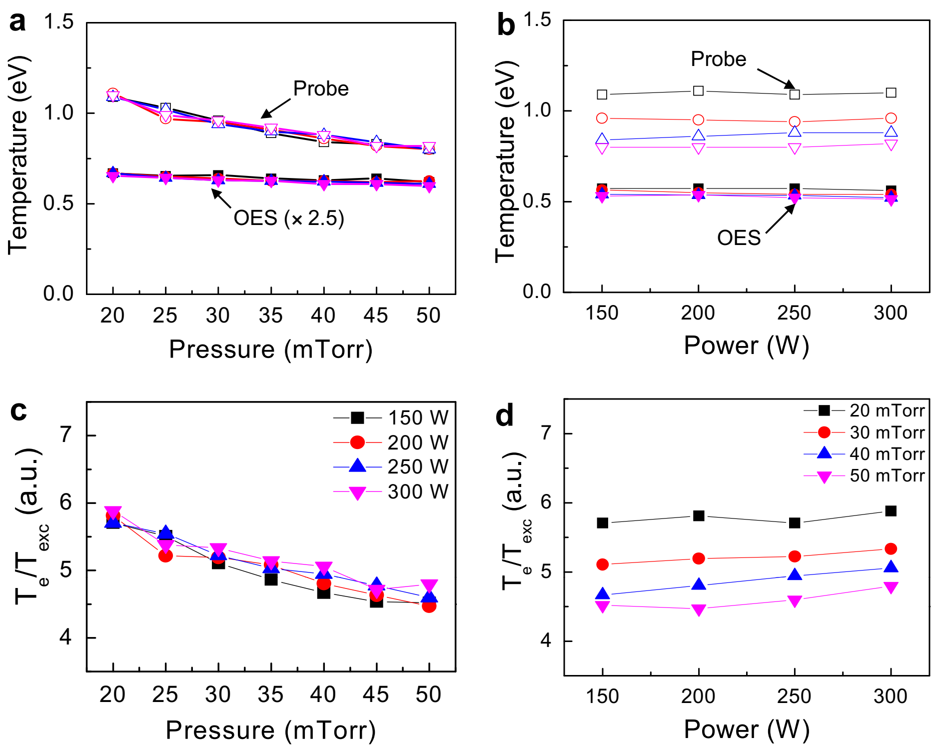

Fig. 4. The two temperatures as a function of (a) gas pressure (20 mTorr–50 mTorr) and (b) input power (150 W–300 W) in MDCP, and their ratios are plotted in (c) and (d).

Fig. 3 shows T_{exc} and T_e and their ratios T_e/T_{exc} measured in an ICP in the range of (1–30) mTorr and (200–800) W. Both temperatures T_{exc} and T_e are found to decrease with the increase in pressure whereas they are only slightly dependent on power. An increase in the electron density with increasing pressure at a fixed power and with increasing power at a fixed pressure was observed, and the values ranged from 1.2×10^{10} to 3.2×10^{11} cm^{-3} in our experiment. Since the electron density increases, the electron temperature decreases at a fixed power. This phenomenon is reflected in Fig. 3(a) and (b). In the high power inductive mode in our experiment, EEPFs exhibited a Maxwellian distribution and T_e obtained by integrating the EEPF using Eq. (6) showed stronger pressure dependence but a weaker power dependence. Similar observations were previously reported in argon ICPs [18,19].

As shown in Fig. 3(c), the T_e/T_{exc} ratio increases to about 3–4 up to 10 mTorr, and thereafter it remains almost constant. The ratio also remains the same as 1.7 (1 mTorr) and 3.8 (15 mTorr and 30 mTorr) from 200 W to 800 W, in Fig. 3(d). These imply that, under a certain condition (10–30 mTorr gas pressure and 200 W–800 W power), it is shown that T_{exc} diagnostics is an indirect method for measuring T_e by multiplying T_{exc} by the ratio value.

In Fig. 4, the two temperatures and their ratios are plotted in (20–50) mTorr and (150–300) W in MDCP. Once the argon discharge is made between the cathode and anode, Ti atoms emitted from the titanium target by argon cation sputtering are diffused to the substrate. The substrate was negatively floating; thus, the electrons are trapped between the cathode and the substrate sheath where both the OES (Ti I) and probe diagnostics were employed (Fig. 1 in ref. [14]). As shown in Fig. 4(b), both T_{exc} and T_e are independent of the input power, as in ICP and CCP cases, at least under this certain pressure and power range. Fig. 4(c) and (d) demonstrate that the temperature ratio tends to decrease linearly from 20 mTorr to 50 mTorr and remains constant from 150 W to 300 W. Therefore, as in CCP and ICP experiments, T_{exc}

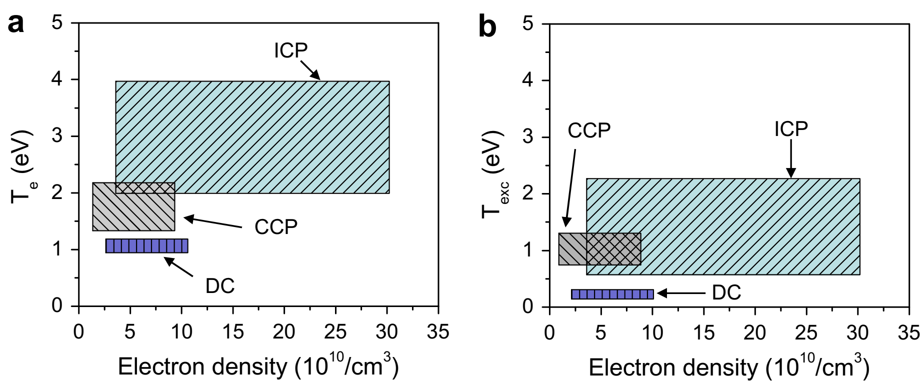

Fig. 5. The measured ranges of (a) T_e and (b) T_{exc} as a function of electron density in various plasmas.

measurement can also be regarded as an alternative diagnostic for T_e in MDCP.

The measured electron and excitation temperatures given in Figs. 2–4 are summarized as a function of measured electron density in Fig. 5. The electron density range is from 2.5×10^{10} cm^{-3} to 1.1×10^{11} cm^{-3} for a DC plasma, 5.2×10^{9} cm^{-3} to 8.3×10^{10} cm^{-3} for a CCP, and 4.0×10^{10} cm^{-3} to 3.2×10^{11} cm^{-3} for an ICP. The areas in Fig. 5 indicate the difference between the absolute maximum and minimum values of the temperatures under the electron density scope. It also demonstrates the electron density and temperature range in which this method is valid.

4. Conclusions

Optical emission from either Ar or Ti was used to determine the electron impact excitation temperature T_{exc} in CCP, ICP, and MDCP, and it was compared with the electron temperature T_e derived from the electron energy probability function obtained using a Langmuir probe. The decrease of T_{exc} and T_e against the increasing gas pressure was observed as well as a weak dependence on the input power.

The measured excitation temperatures were in the range of (0.5–2.3) eV in ICP, (0.6–2.7) eV in CCP, and (0.5–0.7) eV in MDCP, respectively. These temperatures are lower than the measured electron temperatures in (2.1–4.0) eV in ICP, (1.3–2.6) eV in CCP, and (0.8–1.1) eV in MDCP, respectively. The fact that $T_{exc} < T_e$ is attributed to deviation of the plasma from the local thermodynamic equilibrium (LTE) [11].

The present results suggest that measuring T_{exc} with a pre-measured T_e/T_{exc} ratio through a calibration experiment can estimate the absolute value of T_e under the conditions demonstrated in Fig. 5. It also indicates that T_{exc} measurement can be a good alternative diagnostic for various discharges.

Acknowledgments

This work was supported partially by National R&D Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education, Science and Technology (2009-0082624) and partially by Samsung LCD Business, Korea. The authors would like to thank Dr. S. Y. Moon for valuable discussions and Drs. J. H. In and S. K. Ahn for their experimental help, especially in Langmuir probe measurements.

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