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Correlation between excitation and electron temperature in 50 Hz pulsed Ar–O_2 mixture plasma

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ABSTRACT

Optical emission spectroscopy is employed to investigate the excitation temperature T_{exc} and electron temperature T_e in 50 Hz, pulsed DC Ar–O_2 mixture plasma. The excitation and electron temperatures are measured as function of current density, pressure and Ar concentration in the mixture. Boltzmann plot method is employed to estimate the excitation temperature T_{exc} whereas electron temperature T_e is calculated by using modified Boltzmann plot technique. The results show that both T_e and T_{exc} have similar trend i.e., both decreasing with increase in pressure while increasing trend is observed with increase in argon concentration in the mixture. On the other hand small variation is noted regarding change in current density. The results of correlation between electron and excitation temperature suggests that T_e can be measured indirectly from T_{exc} if the variation in ratio T_e/T_{exc}, as function of discharge parameters, has similar trend.

1. Introduction

Oxygen discharges are mostly used for industrial-materials processing such as: dry etching of a photoresist, synthesis of metal oxide, functionalization of polymers, deposition of oxide films, and ashing of samples [1,2]. The experimental investigations of O_2 and their mixtures like O_2/Ar have importance because the addition of Ar to O_2 discharges enhances the stability and maintaining the plasma compositions over a wide range [3]. The different rate processes like: excitation, ionization and dissociation in plasma are mainly electron temperature (T_e) dependent and it also play a key role in chemical process in plasma. On the other hand the unwanted occurrence during processing such as; notching and charge buildup in the substrate of plasma etchers can also be reduced by lowering the electron temperature [4]. 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 [5]. Local thermodynamic condition is not generally achieved in low pressure plasma and such plasmas are called non-LTE plasma. In case of local thermodynamics 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} equals to T_e [6]. In case of laboratory or industrial plasma it is difficult to expect $T_e = T_{exc}$ because they do not satisfy the LTE condition and falls in corona regime thus it is difficult to find T_e spectroscopically. Under such non-LTE plasma, usually Boltzmann plot method is employed to measure excitation temperature whereas modified Boltzmann plot method is used to calculate electron temperature spectroscopically [7].

The purpose of this work is to study the correlation between the excitation temperature and the electron temperature as function of discharge parameters like Ar percentage in the mixture, gas pressure and input current density. Optical emission spectroscopy (OES) is employed to measure these parameters because it is non-intrusive technique that can monitors the changes in discharge plasma accurately over wide range of parameters.

2. Experimental setup

The schematic diagram of the experimental set up is shown in Fig. 1. Two parallel plates in the form of electrodes were inserted in a stainless steel chamber of 62 cm height and diameter. A locally made 50 Hz pulsed DC power supply was attached to the top electrode via inductive load. The side and back of the electrodes were

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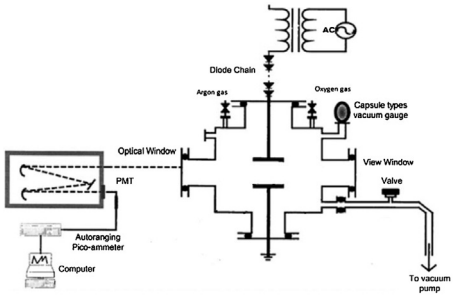

Fig. 1. Schematic diagram of experimental setup.

covered by the ceramic to avoid additional discharge. The chamber had eight main circular multipurpose ports each having diameter of 11 cm. One of the port serve as view window to monitor the state of the plasma generated inside the chamber. The pressure in the chamber is maintained using a rotary vane pump and was recorded with a capsule type pressure gauge. The flow rate of argon and oxygen was noted by a mass flow meter. Time integrated emission spectrum were recorded as function of discharge parameters and then normalized for the spectral response of the: spectrometer, optical fiber, optical lens and quartz window provided by the manufactures. The data were recorded for constant gas flow rate of 50 SCCM, while the filling gas pressure was varied from 1 to 7 mbar. Similarly the current density was also varied in steps like: 0.67, 1.34, 2.01, 2.68, 3.35 mA cm^{-2}.

3. Boltzmann plot

To calculate excitation temperature T_{exc} it is assumed that the upper energy levels of the chosen atomic transitions are in local thermodynamic equilibrium (LTE) that is; the population density of such levels follows the Boltzmann distribution. Taking into account the degeneracy of each atomic state, the population is given by [8]

$$\frac{n_i}{n_j} = \frac{g_i}{g_j} \exp\left(-\frac{E_i - E_j}{KT_{exc}}\right) = \frac{n}{Z} \exp\left(\frac{-E_i}{KT_{exc}}\right) \quad (1)$$

where n is the particle density; n_i and n_j are the number densities of the atoms in the excited levels i and j respectively, E_i and E_j are the excitation energies. g_i & g_j are the statistical weights; K is the Boltzmann's constant, T_{exc} is the excitation temperature, and Z is the atomic partition function. From Eq. (1) and by using knowledge of the populations of two or more states; we can determine the excitation temperature. The population of a state can be determined from the optical emission spectrum; each atomic line can be identified with a transition from an upper energy state i to a lower one j. The line emission intensity related with the transition is proportional to the: number density of the atoms in the upper state, the probability of the transition A_{ij} and the energy of the emitted photon as

$$I_{ij} \propto h\nu_{ij}A_{ij}n_i \propto \frac{h}{\lambda_{ij}}A_{ij}g_i \exp\left(-\frac{E_i}{KT_{exc}}\right) \quad (2)$$

Eq. (2) can be expressed as straight line equation;

$$\ln\left(\frac{I_{ij}\lambda_{ij}}{A_{ij}g_i}\right) = -\left(\frac{E_i}{KT_{exc}}\right) + C \quad (3)$$

Excitation temperature can be calculated from the slope of this line by using number of Ar–I lines. Here λ_{ij} is the wavelength, I_{ij} is the emission intensity and C is the constant for given species. The spectroscopic data used for the measurement of excitation temperature are given in Table 1.

Table 1
Ar–I spectral lines used to measure the excitation and electron temperature.

λ_{ij} (nm)	g_i	E_i (eV)	$A_{ij(s^{-1})}$	$\sum_{i>j} A_{ij}\left(10^6\ s^{-1}\right)$	No of transitions
516.22	3	15.30	1.9×10^6	34.57	3
693.76	1	14.69	3.0×10^6	5.64	4
706.70	5	13.30	3.8×10^6	34.46	4
731.17	3	14.85	1.7×10^6	5.58	8
750.38	1	13.48	4.4×10^7	44.70	2
751.46	1	13.28	4.0×10^7	40.20	1
763.51	5	13.17	2.4×10^7	34.40	3

4. Modified Boltzmann plot

In plasma parameters electron temperature (T_e) has a vital role because the electron temperature (T_e) gives the knowledge of; collisional process generating active species, density of active species and reactions in the plasma. Presently the understudy plasma belongs to non-LTE plasma and falls in coronal regime in which electron impact excitation is forward process whereas spontaneous decay is reverse process. In such plasmas Boltzmann plot method gives excitation temperature only. In order to determine the electron temperature (T_e) spectroscopically the modified Boltzmann plot technique is employed which is based on corona balance. In this method; several spectral lines with various excitation energies are selected and their spectral intensities are used for the determination of the electron temperature. Since the under study plasma falls in the corona balance regime, hence it is assumed that the levels are populated according to the following corona balance Eq. [6].

$$n_e n_1 X_{1i} = n_i \sum_{j<i} A_{ij} \quad (4)$$

where n_i, n_1 and n_e are the excited state population, ground level and electron densities respectively. $\sum A_{ij}$ is the accumulative transition probability for spontaneous emission from the higher level i to all the other lower levels j, X_{1i} is the rate coefficient regarding electron impact excitation from the ground level 1 to level i. The density of excited level n_i can be written as

$$I_{ij} \propto h\upsilon_{ij}A_{ij}n_i \quad (5)$$

For the first approximation, if we assume that the electron energy distribution function (EEDF) for the free electrons in plasma is Maxwellian then we can write the approximate electron impact excitation rate from the ground state of argon as given by Drawin [9]

$$X_{1i} = 8.69 \times 10^{-8} \times \alpha_{1i} \times z_{eff}^{-3} f_{1i} \times \frac{U_a^{3/2}}{U_{1i}} \times \Psi_a\left(U_{1i}, \beta_{1i}\right)\ cm^3\ s^{-1} \quad (6)$$

where $U_a = 13.6\,kT_e$, $U_{1i} = (E_1 - E_i)/kT_e$, α_{1i} is a constant with a value approximately equal to 1, $Z_{eff} = Z - N + 1$ is the effective atomic number (with Z and N being the atomic number and the number of bound electrons, respectively) and f_{1i} being the absorption oscillator strength of the transition $1 \rightarrow i$ [10].

For atomic species $Z_{eff} = 1$. The function $\Psi_a\left(U_{1i}, \beta_{1i}\right)$ is [9]

$$\Psi_a\left(U_{1i}, \beta_{1i}\right) = \frac{\exp(-U_{1i})}{1 + U_{1i}} \times \left[\frac{1}{20 + U_{1i}} + \ln\left(1.25 \times \left(1 + \frac{1}{U_{1i}}\right)\right)\right] \quad (7)$$

where $\beta_{1i} \approx 1$.

The value of the electron impact excitation rate coefficient X_{1i} can be calculated from Eq. (6) for different values of T_e because it is strongly dependent upon electron temperature T_e. Now it becomes