Spectroscopic measurements of the electron temperature in low pressure microwave 2.45 GHz argon plasma

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Abstract

The main goal of this work is to obtain the plasma electron temperature T_e by optical emission spectroscopy of low pressure microwave argon plasma, as a function of working pressure and microwave power. A plasma system was designed and constructed in our laboratory using a magnetron of domestic microwave oven with power 800W without any commercial part. The applied voltage on the magnetron electrical circuit is changed for the purpose of obtaining the variable values of the microwave power. The spectral detection is performed with a spectrometer of wavelength range (200−1000nm). The working pressure and magnetron applied voltage were 0.3-3.0mbar and 180-240V, respectively. Two methods had been applied to estimate the electron temperature, the ratio of two lines' intensity and Boltzmann plot method. It was found that, for the plasmas investigated, an increase of the electron temperature when the applied voltage has been increasing, while the electron temperature decreases when the working pressure is increasing.

قياسات طيفية لدرجة حرارة الالكترون في بلازما الاشعة المايكروية 2.45GHz لضغط الأركون المنخفض

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الخلاصة

الهدف الرئيس من هذا العمل هو حساب درجة حرارة الكترونات البلازما بواسطة طيف الانبعاث الضوئي لبلازما الاشعة المايكروية لضغط الأركون المنخفض كدالة لضغط العمل وقدرة الاشعة المايكروية. تم تصميم وبناء منظومة البلازما في مختبرالبلازما (قسم الفيزياء، كلية العلوم، جامعة بغداد) باستخدام مجهز اشعة مايكروية (ماكنترون) المستخدم في فرن الميكروويف المنزلي وبقدرة 800W و بدون أي جزء تجاري. لغرض الحصول على قيم قدرة اشعة مايكروية متغيرة يتم تغيير الجهد المطبق على الدائرة الكهربائية للماكنترون. اجريت قياسات طيفية بمطياف مدى الطول الموجي له (200-1000nm). وكان مدى ضغط العمل والجهد المطبق على الماكنترون (0.3-3.0mbar) و (180-240V) على التوالي. استخدمت طريقتين لحساب درجة حرارة الإلكترون، نسبة الشدة لخطين وطريقة مخطط بولتزمان. وقد وجد أن درجة حرارة إلكترونات البلازما تزداد مع زيادة الجهد المسلط، في حين تنخفض درجة حرارة الإلكترونات عند ازدياد ضغط العمل.

Introduction

A glow discharge generated by a microwave power applied through coupling of electromagnetic radiation with frequency ranging (0.3-10) GHz is named as microwave induced plasma[1]. Power supplies working with a frequency of 2.45GHz are common use in industry and domestic microwave oven makes suitable power source for microwave plasma generation [2]. This type of plasma may be considered in local thermodynamic equilibrium (LTE) and non-local thermodynamic equilibrium (non-LTE) depending on the plasma pressure and the electron number density [3]. In the plasma have a low electron density only a small amount of collisions occur and resultantly plasma species are characterized by different temperatures due to insufficient collisional transfer of energy among them. Thus electron number density plays an important role in differentiating the LTE and non-LTE plasma regimes. There are many applications of non-LTE plasmas such as deposition of thin films, etching and cleaning. Among many of the plasma species, electrons have higher thermal velocities due to their lighter mass and therefore can be accelerated by the electromagnetic radiation as compared to other plasma species. These energetic electrons in turn may excite or ionize the plasma species having energy of several electron volts above their ground state by inelastic collisions. Therefore, electron temperature T_e and electron number density n_e are important parameters for describing the plasma discharges as a function of discharge conditions. Moreover, in many cases, the rare gases are used as a feed gas or added as a trace gas in low pressure discharges, and the spectral lines intensities of their optical emission is used to determine these parameters

intrusively [4, 5]. Mainly, argon mixed reactive plasmas is widely used in different material processing applications [6, 7].

Argon plasmas have been widely used in material processing and surface modification applications, because argon is an inert gas and has an appropriate atomic mass among noble gases [8]. Knowledge of plasma parameters, such as electron temperature, electron number density play a key role in the understanding of physics and chemistry of argon plasma for better optimization of processing reactor [9].

In the plasma processing, elementary processes such as ionization, dissociation, and excitation as well as ion bombardment are closely related to the plasma parameters and therefore, plasma diagnostics are important in understanding the mechanism of material processing applications [10]. There are a lot of researches working on microwave induced plasmas and investigation of the plasma parameters and discharge conditions [11-13]. However, a little investigation was carried out on correlation of plasma parameters with discharge conditions in microwave induced plasma via spectroscopy diagnostics.

Optical emission spectroscopy is an appropriate and inventive technique to determine the plasma parameters such as electron temperature, electron number density and EEDF [14]. The electron temperature in low pressure plasma may be extracted from the emission spectrum of atomic and molecular species that are excited by direct electron impact process and depopulated by the spontaneous emission of radiation [15]. Several spectroscopic methods are being used to determine the electron temperature T_e including lines-ratio method and

Boltzmann plot method [13]. These two methods adopted here to determine the electron temperature. This work, an optical emission spectroscopy (Thorlabs CCS200, wavelength range 200-1000nm) to observed plasma emission lines and measuring of the electron temperature of microwave argon plasma discharge versus filling pressure at different input powers.

Microwave power consumed in the plasma reactor can't be measuring due to a lack of necessary equipment to measure it; for this reason the term "applied voltage" was considered instead of microwave power in this measuring. This approximation is possible because the microwave power consumed is a function of the applied voltage on the electrical magnetron circuit, thus the applied voltage is considered in the measurements below.

Electron temperature determination by optical emission spectroscopy technique

Optical emission spectroscopy (OES) is the most popular technique to investigate gas discharge (plasma). Since it is a simple technique and that it produces no perturbation in the plasma. There are four plasma models dependent on the mechanism of electron interaction. These are the local thermal equilibrium (LTE) model, the steady state Corona model, the time dependent Corona model, and the collisional radiative (CR) model [16].

The methods used to measure the plasma parameters depend on the plasma model. Three methods are available for electron temperature measurement, these are [16]:
1) The ratio of two lines' intensity.
2) The ratio of a line to continuum intensity.
3) The ratio of two parts of continuum intensities.
The simplest and most direct method of using spectral line intensities to

determine the temperature of plasma is to use the relations existing between the intensities of the spectral lines, when the atomic state densities are in equilibrium. In order to determine the electronic plasma temperature by using the relative intensities between two spectral lines, it is necessary to consider plasma in the local thermodynamic equilibrium (LTE). The LTE basically demands that the excitation and ionization processes have been produced only by electron impact. To satisfy this condition, it is necessary for the electronic plasma density and temperature to be $n_e \geq 10^{14}$ m^{-3} and $T_e < 1$ eV, respectively. In our work these conditions were satisfied for "cold" electrons.

The electron temperature can be estimated by using the intensity ratio of a couple emission lines labeled 1 and 2 in flowing equation [17]:

$$\frac{I_1}{I_2} = \frac{A_1 g_1 \lambda_2}{A_2 g_2 \lambda_1} exp\left(-\frac{E_1 - E_2}{kT_e}\right) \qquad (1)$$

where I_1, I_2 are the intensities of two spectral lines, A_1, A_2 are the transition probability of two spectral lines, λ_1, λ_2 are the wavelength of two emission lines, g_1, g_2 are the statistical weights of two spectral lines, E_1, E_2 are the upper level energies of the transitions of the producing two spectral lines and k is the Boltzmann constant. Extracting T_e from Eq. (1):

$$kT_e = \frac{E_1 - E_2}{Ln\left(\frac{I_2}{I_1}\right) - Ln\left(\frac{A_2 g_2 \lambda_1}{A_1 g_1 \lambda_2}\right)} \qquad (2)$$

By using the Eq. (2); the electron temperature has been calculated. The values of E_1, E_2, A_1, A_2, g_1 and g_2 are obtained from [18].

Also, the plasma electron temperature T_e can be calculated using the Boltzmann plot method. The Boltzmann plot method is an uncomplicated and broadly used

method for spectroscopic measurement, mainly for measuring the electron temperature of plasma from using the relative intensity of a several spectra having relatively large energy differences [19]. However, in order to practically apply the Boltzmann plot method for the measurement of electron temperature, the excitation level needs to be reached under a LTE condition.

Boltzmann plot method consists of measuring the relative intensities of several emission lines [20]. It's can be described below, consider two energy levels, E_i and E_j (E_i is the lower level and E_j is the upper level), of two different line intensities with respective atomic densities of N_i and N_j. Under the condition of thermal equilibrium, the relation between N_i and N_j can be established using the Boltzmann distribution:

$$\frac{N_j}{N_i} = \frac{g_j}{g_i} exp\left[-\frac{E_j - E_i}{kT}\right] \qquad (3)$$

where g_i, g_j are the statistical weights of the respective states, k is the Boltzmann constant (1.38×10^{-23} J/K), and T is the temperature in K. If the total population density is N, the Boltzmann relation valid to the population distribution over a given atomic state is given by:

$$\frac{N_j}{N} = \frac{g_j}{Z(T)} exp\left[-\frac{E_j - E_i}{kT}\right] \qquad (4)$$

where $Z(T)$ is the sum of the weighted Boltzmann function of all the discrete energy levels, which is the partition function and it's given by:

$$Z(T) = \sum_m g_m \, exp\left[-\frac{E_m}{kT}\right] \qquad (5)$$

but, when the plasma atom is de-excited from the upper energy level, E_j, to the lower energy level, E_i, the optical intensity of spectral line can be expressed by the following equation:

$$I_{ji} = \frac{hc_0}{4\pi\lambda_{ji}} A_{ji} N_j \qquad (6)$$

where λ_{ji} is the wavelength of the emitted light, h is the Planck's constant (6.626×10^{-34} J. s), c_0 is the velocity of light in vacuum (3×10^8 m/s), and A_{ji} is the transition probability, which is the probability per second that an atom in state j spontaneously emits in a random direction and is de-excited to state i. From Eq. (6) and Eq. (4) and rearranging they give:

$$\frac{I_{ji} \lambda_{ji}}{A_{ji} \, g_j} = \frac{hc_0 N}{4\pi Z(T)} exp\left[-\frac{E_j}{kT}\right] \qquad (7)$$

by taking the logarithm in both sides of Eq. (7) obtained:

$$Ln\left(\frac{I_{ji} \lambda_{ji}}{A_{ji} \, g_j}\right) = -\frac{E_j}{kT} + C \qquad (8)$$

where C is a constant. Now, by plotting the term $Ln\left(\frac{I_{ji} \lambda_{ji}}{A_{ji} \, g_j}\right)$ in the vertical axis with E_j in the horizontal axis from Eq. (8) for several lines (all of these lines have the same lower energy level), the electron temperature T_e which is related to the slope of the linear fitting [21-23].

Experimental work

The plasma chamber was constructed in our laboratory. The main part of plasma reactor was built from a stainless steel 316L ultra high vacuum (UHV) T-pipe (T-LF100, thickness 4 mm, inside diameter of flange is 100mm and outside diameter is 130mm). Two stainless steel 316L flanges with diameter 130mm and thickness 8mm and centrally drilled with 24mm was fabricated to connect the sides of main chamber. A quartz tube with outer diameter of 23mm is located at the axial chamber. The microwaves are coupled into the vacuum chamber in the form of a coaxial waveguide, consisting of an inner copper rod and a surrounding the quartz tube with atmospheric pressure between them (quartz tube and copper pipe). The plasma is produced outside the quartz tube.

The microwaves are guided from the generator (Magnetron) through rectangular waveguides into the vacuum vessel. The inner rod (copper pipe) serves as an antenna for the microwaves, but the outer is cylindrically shaped dielectric tube (quartz). The quartz tube is vacuum sealed and forms a part of the large vacuum vessel. Consequently the plasma ignites around the quartz tube in the low pressure volume. That

implies that a surface wave can propagate at the interface between dielectric tube and plasma. The schematic diagram of the plasma system parts is shown in Fig. 1. The vacuum system consists of a rotary pump followed by an adsorption pump. It can be maintained at pressures ranging from atmospheric pressure to $<10^{-3}$ mbar. For the accurate measurement of a pressure inside the plasma chamber, pirani gauge is used.
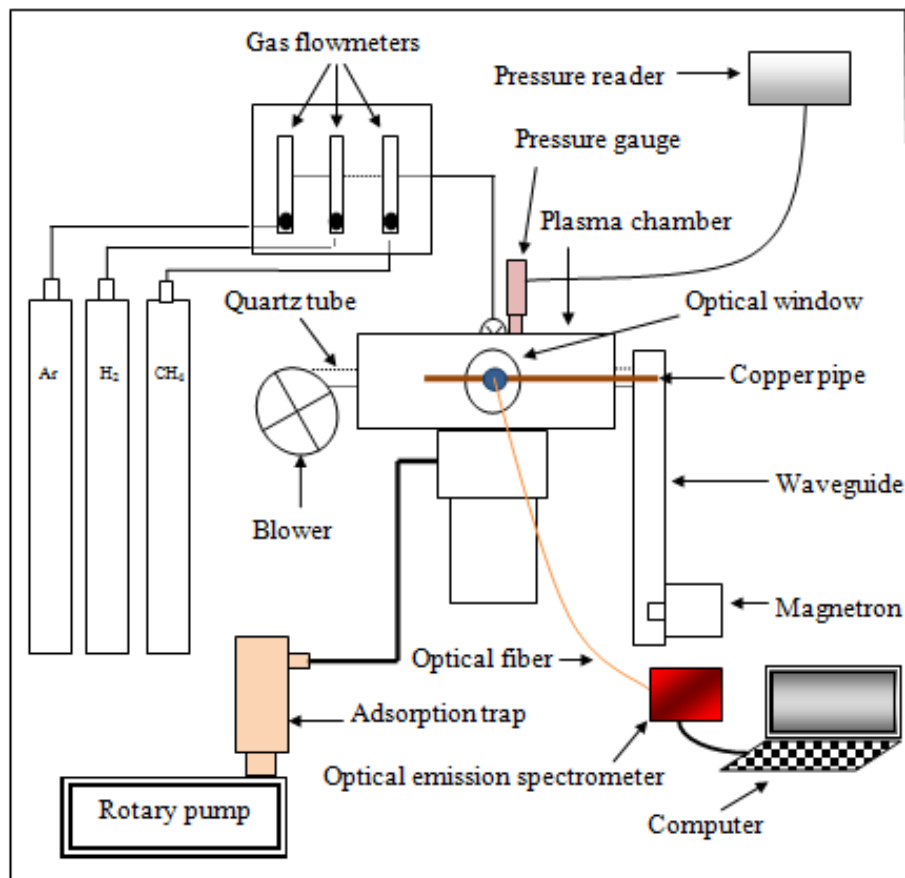

Fig.1: Schematic diagram of plasma system.

Results and discussions

Spectra of microwave discharge were recorded in the wavelength range 350-950nm as shown in Fig. 2. They were mainly composed of neutrals lines in the spectral region 690-930nm due to

losses in the power coupling from the magnetron to the plasma chamber. The first method (The ratio of two lines' intensity) was used to estimate the electron temperature in the argon microwave discharge.
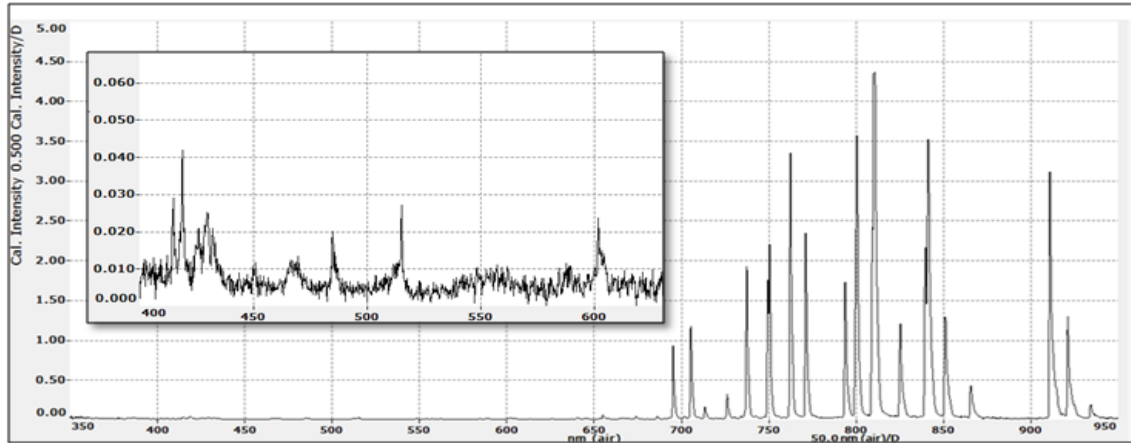
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Fig.2: Emission spectra in microwave argon plasma at pressure 0.3mbar and applied voltages is 180V.

Determination of electron temperature T_e using optical emission spectroscopy technique by estimating the ratio of the intensity of two lines has been adopted here. For high accuracy, it is desirable to choose two spectral lines, whereas the ratio of the relative intensity of the two lines is a strong function of T_e. This will be the case if the difference between the transition energy levels of the two lines is a large enough [17]. The accuracy may be improved somewhat by measuring spectral lines for several times (here every spectrum is repeated three times) and averaging the resulting temperatures. The two lines (750.38, 810.369)nm are chosen to calculate T_e when the pressure values were 0.3, 0.5, 1.0, 2.0 and 3.0 mbar and the magnetron applied voltage were 180, 190, 200, 210, 220, 230 and 240 V. Table 1 lists the values of the parameters E, A and g for the two lines Ar I (750.38) nm and Ar I (810.36)nm have been taken from [18].

Table 1: Values of transition probability, upper energy level and statistical weight that used to calculate T_e [18].

Wavelength (nm)	Transition probability A, (s^{-1})	Upper energy level E, (eV)	Statistical weight g
Ar I (750.38)	4.45×10^7	13.47	1
Ar I (810.36)	2.5×10^7	13.15	3

Using Eq. (2) and substitute values of A, E and g from Table 1 the electron temperature T_e can be calculated, these values are tabulated in Table 2.

Table 2: Electron temperature T_e (eV) at different working pressures (mbar) and applied voltages (V).

Applied Voltage (V)	Working pressure (mbar)				
	0.3	**0.5**	**1.0**	**2.0**	**3.0**
	Electron temperature T_e (eV)				
180	0.38	0.36	0.29	0.23	0.20
190	0.44	0.42	0.37	0.26	0.22
200	0.50	0.47	0.43	0.29	0.25
210	0.55	0.51	0.47	0.31	0.27
220	0.58	0.54	0.49	0.33	0.28
230	0.62	0.57	0.54	0.34	0.29
240	0.62	0.61	0.57	0.36	0.30

Fig.3 shows the variation of spectral lines intensities for Ar I (750.38)nm at different applied voltage and filling pressures. The intensity of spectral lines increases when the applied voltage is increasing and decreases when the pressure is increasing. By increasing of the pressure, the neutral density increases, the mean free path of the electron-neutral impacts excitations decreases, and the frequency of these collisions are increasing. By increasing the power, the ionization degree, as well as the electron energy increases, so the electron-neutral impact excitation rate will increase. The electron transfers their energy by impact with other particles and excites them. By de-excitation, atoms and ions emit spectral lines.
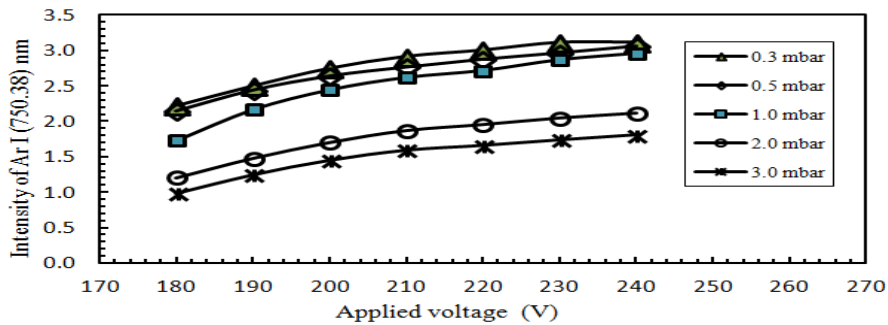

Fig.3: Line intensity of Ar I (750.38)nm (a. u.) as a function of applied voltage (V), at different pressures (mbar).

Fig.4 represents the values of electron temperature T_e (eV) that tabulated in Table 2 as a function of applied voltage at different values of pressures. Also, Fig. 5 shows these values of electron temperature T_e plotted as a function of pressure at different applied voltages (V).

Figs.4 and 5 show the decreasing of T_e with the increase in gas pressure may be explained as follows: when the gas pressure increases, it causes an increase in the number of collisions bet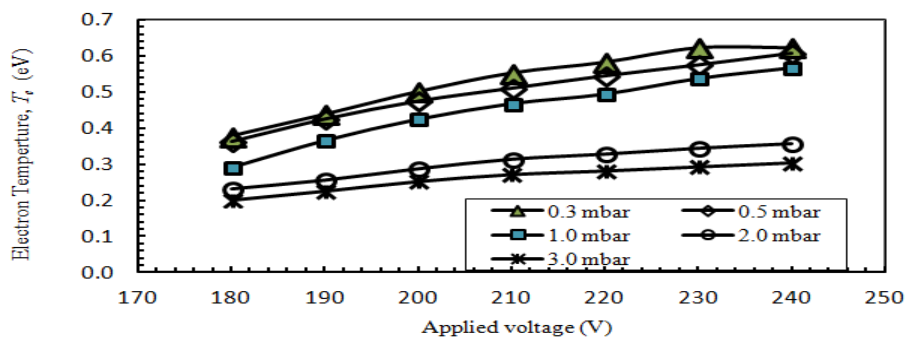ween the electrons and the gas atoms. As a result the energy transferred from the electrons to the gas particles increases causing an increase in the gas temperature by lowering the electron temperature. Also, the mechanism of excitation and ionization of atomic and ionic species in argon plasma is supposed to occur mainly by electron impact. When the filling pressure is increased, the high-energy tail of the electron energy distribution function contracts to the lower energies. Therefore the ionization, which results from the energetic electron's impact with gas atoms, is reduced.

Fig.4: Electron temperature T_e (eV) as a function of applied voltage (V), with different pressures.
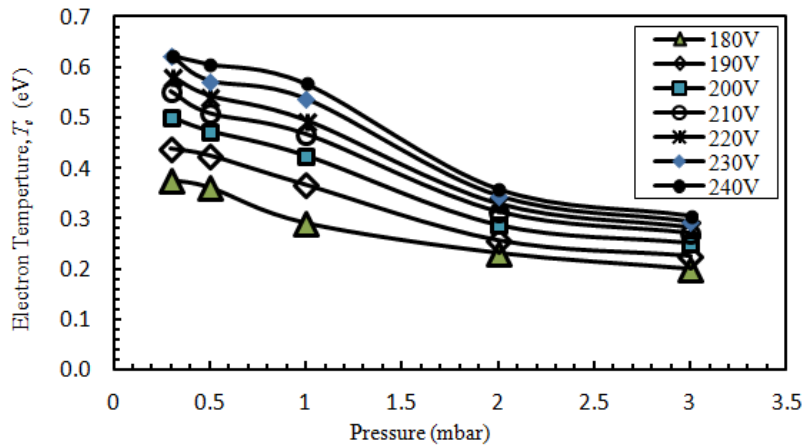
Fig.5: Electron temperature T_e (eV) as a function of pressure (mbar), with different applied voltages (V).

Also, electron temperature T_e can be determined using Boltzmann plot method. In this method, several spectral lines are considered (represent Ar I); all of these lines have a similar lower energy level E_i with different upper energy level E_k. In this study, seven lines have been taken (415.85, 696.54, 706.72, 763.51, 772.37, 801.47 and 811.53) nm. Table 3 lists argon lines considered with their spectroscopic data.

Table 3: Spectroscopic data of argon lines used for OES diagnostic [18].

Ion	Wavelength (nm)	$A_k \times 10^6 (s^{-1})$	E_i (eV)	E_k (eV)	g_k
Ar I	415.85	1.4	11.54	14.52	5
Ar I	696.54	6.39	11.54	13.32	3
Ar I	706.72	3.80	11.54	13.30	5
Ar I	763.51	24.5	11.54	13.17	5
Ar I	772.37	5.18	11.54	13.15	3
Ar I	801.47	9.28	11.54	13.09	5
Ar I	811.53	33.1	11.54	13.07	7

Using Eq. (8) and substitute values A_k, E_k and g_k from Table 3 for seven argon lines $Ln\left(\dfrac{I_{ji}\,\lambda_{ji}}{A_{ji}\,g_j}\right)$ can be plotted as a function of E_j, the electron temperature T_e is related to the slope of the linear fitting. Electron temperature T_e can be calculated for different pressures and different applied voltages. Fig. 6 represents the electron temperature T_e (eV) as a function of applied voltage (V) at different pressures obtained by Boltzmann plot method).

Fig.6 shows that, there is increasing in electron temperature T_e when applied voltage is increasing at the same pressure. While, T_e is decreasing when the pressure is increasing at the same applied voltage. This behavior is similar to that obtained from Figs. 4 and 5.
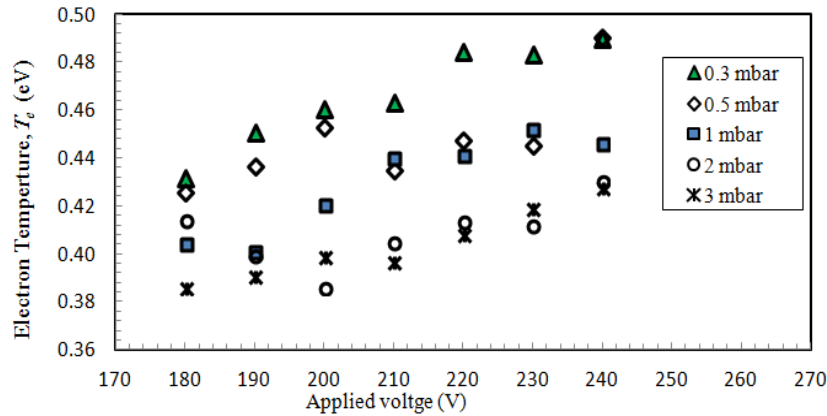
Fig.6: Electron temperature T_e (eV) as a function of applied voltage (V), at different pressures (using Boltzmann plot method).

Conclusions

The two lines' intensity and Boltzmann plot methods can be used successfully to determine the electron temperature in a variety of microwave power and low pressure argon plasmas. Basic motivation of this study is to explore the effects of filling pressures and microwave powers on the excitation and ionization process involved in optical emission. Experimental results show that the electron temperature increases when the applied voltage is increasing and the pressure is decreasing. Through the comparison between the two above methods and according to the results that we had obtained, it's found that the ratio of two lines' intensity method is the most convenient in such measurements.

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