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Electron Temperature measurements Te spectroscopically in argon pulsed cylindrical hollow cathode plasma jet

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ABSTRACT

The properties of low pressure argon pulsed hollow cathode discharge were investigated using Rogoviski coil, and time resolved Optical emission spectroscopy (OES) under different conditions of gas flow rate (20-240 sccm), and at charging voltage 4kv. The Electron temperatures were measured in argon plasmas as a function of time, using the argon line emission intensity ratio of 4675A^0/4259A^0 and 4348A^0/4159A^0. Also the electron temperature was determined by adding small admixture of helium gas (5sccm) to the working argon gas without disturbing the plasma properties, using the intensity helium line ratio He 4713A^0/4921A^0 and 5048A^0/4713A^0. The electron variation density with time was measured using the line intensity of exited argon ion 4880A^0. The results obtained indicated that the maximum discharge current is approximately 6.7kA. It was found that the electron temperature decreases with the increase of gas flow rate; this is attributed to the increase in the number of collisions between electrons and another plasma species.

INTRODUCTION

The pulsed hollow cathode plasma discharge was investigated as a source plasma jet for deposition of different thin films. A pulsed discharge is low-pressure high voltage current characterized by the presence of an axial hole in the cathode. Pulsed sputtering allows reducing the substrate temperature during deposition and considerably increasing plasma density in the pulse-on time (Cada, M. *et al*., 2006). The increase in the plasma density can promote formation of more dense and stable film structure that leads to improvement of the film properties. In low pressure cylindrical hollow cathode, the discharge takes place almost entirely inside the hollow cathode. The pressure in the experimental chamber 10^{-2} torr, while the pressure inside the nozzle is always higher than $25X10^{-2}$ torr, so the incoming working gas forces the discharge out of the nozzle into the experimental chamber and plasma jet interact with the substrate (Cada, M. *et al*., 2003). The hollow cathode discharge has high ionization efficiency; high plasma density, bright discharge, and photon yield a spectroscopic measurement.

Optical emission spectroscopy (OES) is based on the measurement of the optical radiation emitted from the plasma as it reflects the properties of the plasma in the immediate environment of atomic, molecular, and ionic radiators (Behringer, K. *et al*., 1994). The radiation is the result of electron or ion interaction with other particles in the plasma. Because of their high velocity, electron interactions tend to dominate the collisional excitation and ionization processes. Three electron transitions may occur during these interactions: bound-bound transitions; bound-free transitions; and free-free transitions. The light emitted as a result of these transitions forms line, band, or continuous spectra (Abdou, A. Garamoon *et al*., 2007).

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Time-resolved emission optical spectroscopy was used to measure the electron temperature and the electron density and to study the dynamics of pulsed plasmas. The advantage of this method is monitoring the discharge without disturbing plasma (Cada, M. *et al*., 2006). A small admixture of helium gas can also be added to plasma without disturbing the plasma properties for the purposes of optical diagnostics

In the present paper, we investigate the intensity variations of the Argon emission lines, which are identified to different transitions when glow discharge source is operated with dc pulsed power supply, in order to compare each plasma conditions. Also, the electron temperature Te in Argon gas at different experimental conditions was determined by two methods using optical emission spectra by the relative intensity line to line ratio method.

Experimental Setup and diagnostic techniques:

The schematic diagram of the experimental setup is showing in figure (1). The plasma source of this experiment has been described in some details in a previous paper (Khaled Hussien Metwaly *et al*., 2016) and will be briefly described here. The hollow cathode is made of metal copper of the cylindrical shape of diameter 25mm to absorb the produced heat by ions sputtering in the cathode. There is a hole of diameter 5mm in the H.C. axis. The anode ring made of copper and has an inner diameter 40mm. The H.C. chamber made from Pyrex glass with diameter of 75mm and height 150mm.

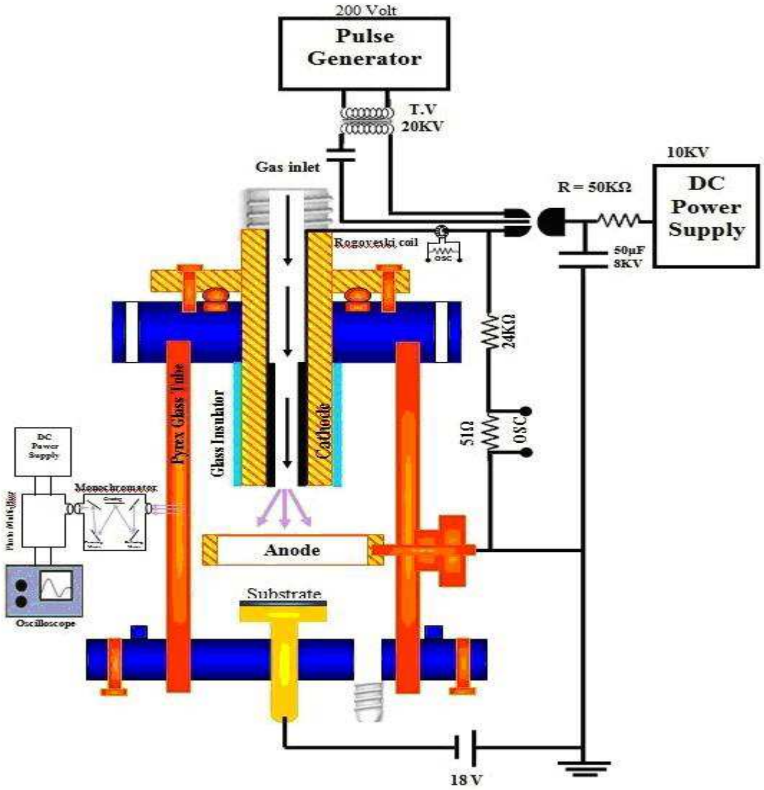

Fig. 1: The schematic diagram of the Hollow cathode plasma Jet

The experimental chamber is continuously pumped by a rotary pump and an oil diffusion pump. The residual pressure of the vacuum apparatus reaches the order of 10^{-4} torr. The gas flow rate to the experimental chamber is controlled by a mass flow rate meter. The discharge can be powered by a negative pulsed DC system. The power system consists of a source of a half wave rectifier voltage 4kV, spark gap switch, condenser bank (50μF, 8kV) and pulse generator of (SCR) circuit. The discharge current was measured by a Rogowski coil.

The intensity of argon emission selected lines from the pulsed hollow cathode discharge was analyzed by Mc Pherson scanning monochromator [Model 270] with 35cm focal length. The entrance slit of the monochromator is at 15 cm from optical window such that sufficient light is transmitted to the slit without light-collecting optics. The emission spectra are recorded as function of time. The wavelength of the monochromator is calibrated using a mercury lamp before the experiment. The emission intensity profiles used for the spectroscopic analysis are normalized for the spectral response of the photo-multiplier tube (type 9558QB).

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RESULTS AND DISCUSSION

3.1. Discharge characterization:

Figure (2) shows the photograph of light emission from pulsed hollow cathode plasma jet at charging voltage 4kV, an argon gas pressure 10^{-2} torr. The photo shows the expanding discharge with the densest plasma at the outlet of the nozzle. The emission plasma of the outlet nozzle works as a positive column of the DC glow discharge (Hubička *et al*., 2003).

Fig. 2: Photograph of light emission from pulsed DC hollow cathode plasma jet

Figure (3) shows time resolved of intensity exited atom line ArI (4159A^0) and discharge current waveform at charging voltage 4kV, Argon pressure 10^{-2} torr. It is remarkable from experimental results that maximum discharge current is approximately 6.7kA. The discharge current is dumped oscillation. This is mainly caused by the inductance in the discharge loop. (Hubička *et al*., 2003). the half period of the discharge current is about 40 µs, and the rise time of discharge current is 20 µs.

Figure (4) shows time resolved of intensity exited ion line ArII (4348A^0) and discharge current waveform at charging voltage 4kv, Argon pressure 10^{-2} torr.

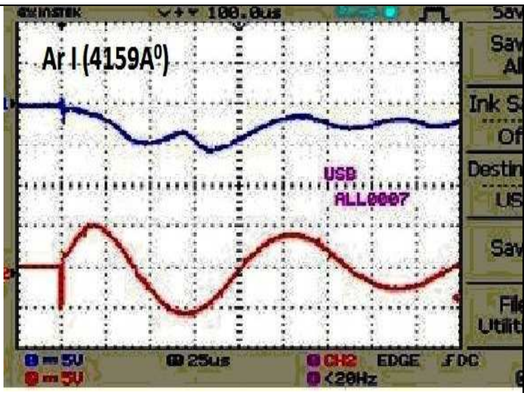

Fig. 3: Time resolved of intensity exited atom line ArI (4159A^0) and discharge current waveform at charging voltage 4kV, Argon pressure 10^{-2}.

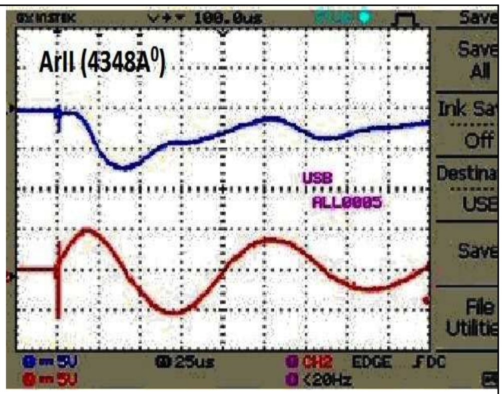

Fig. 4: Time resolved of intensity exited ion line ArII (4348A^0) and discharge current waveform at charging voltage 4kv, Argon pressure 10^{-2} torr.

An excited Ar+ level can be reached by either the one-step process

$$Ar + e^-(E > 35\,eV) \rightarrow Ar^{+*}$$

Or the two-step process

$$Ar + e^-(E > 15.5\,eV) \rightarrow Ar^+$$

$$Ar^+ + e^-(E > 19.5\,eV) \rightarrow Ar^{+*}$$

Since for a typical low-temperature plasma (≈4eV) there are very few electrons with the 35 eV required for the simultaneous excitation and ionization by the one-step process, there is also the possibility of a large signal contribution from the excitation of the ground state ions in the plasma by the two-step process (John B Boffard *et al*., 2004).

Emissions from atomic lines have a similar two-step excitation possibility by means of excitation from metastable atoms present in the plasma. Excitation of metastable states would lead to increased emissions over those predicted from electron-impact excitation of the ground state (Namjun Kang *et al*., 2011).

The ArI and ArII line intensities in the spectra recorded during and after the first discharge current pulse shows that during the discharge current period, the atom and ion excitation is caused mainly by impact with energetic electrons, while at the afterglow the recombination processes play a major role in the ArI and ArII energy levels population (Surmeian, A. *et al*. 2005). The presence of the hump profile in the temporal afterglow of ArI and ArII emission lines is due to recombination processes involving ions generated by metastable - metastable collisions, the atom and ion metastables being the only source of excitation and ionization in the temporal afterglow plasma (Glen, P. Jacksona *et al*., 2003). As shown in figure (3, 4) the after – peak intensity of spectral line is dependent on the discharge energy. It means that in the discharge, the dominant production of ArII is provided by the three body electron- ion recombination because of the inverse of electron temperature and the strong dependent on the electron density.

3.2. Corona Model:

The electron densities and electron temperatures measured by single Langmuir probe in the previous work (Khaled Hussien Metwaly *et al*., 2016) were of the order of 10^{-12} cm^{-3} and 4-10 eV, and, therefore, embrace the regime where the plasma is in Corona model. Here, excitation occurs by electron impact and de-excitation by radiative transitions. In low density plasma, the number density of atoms is low enough to neglect excited-state atom–atom and excited-state ion–atom collisions. Furthermore, in weakly ionized plasma the electron and ion densities are low in comparison with the atomic density. Thus, the primary channels for excitation/de-excitation of atoms are with photons and electrons. In corona equilibrium (Mc. Whirter, R.W.P., 1965), atomic excitation is through electron impact excitation of ground state atoms and de-excitation is through photon decay.

In the steady-state corona model, the plasma is assumed to be optically thin, i.e., the radiation escapes without interacting with the plasma and if there is a change in the plasma parameters, it takes place sufficiently slowly for the population densities to take up their new steady-state values at each instant. A Maxwellian velocity distribution is assumed for the free electrons. The corona model assumes a balance between the collisional ionization and radiative recombination (Podder, N.K. *et al*. 2004).

In corona model, the electron temperature can be determined the intensity ratio of emission lines derived from transitions between levels i-x in the excited singly-charged ion ($I^{+}_{i,x}$) and p-y in the excited atom ($I^{0}_{p,y}$), the electron temperature may be determined all using the following expression (Mc. Whirter, R.W.P., 1965):

$$\frac{I^{+}_{i,x}}{I^{0}_{p,y}} = C.T_e^{0.75}.\exp\left[\frac{-\{E_g + E_{i,g} - E_{p,g}\}}{kT_e}\right] \tag{1}$$

where k is the Boltzmann constant, Te the electron temperature, Eg the ionization potential of the atom, $E_{i,g}$ the excitation energy of the singly charged ion from its ground state (g) to level i, $E_{P,g}$ the excitation energy of the atom from ground state to level p. C is a constant, which depends on oscillator strengths and transition probabilities of the involved transitions.

3.3. Electron temperature of Argon plasma:

Values of the spectroscopic parameters used in equation (1) have been taken from (Podder, N.K. *et al*. 2004). The first method to determine the electron temperatures in argon plasmas as a function of time, using the argon line emission intensity ratio of $4675A^0/4259A^0$, at $4765A^0$ (Ar^+) ($E_{i,g}$ =11.92+14.74=36.66eV), and $4259A^0$ (Ar^0) ($E_{P,g}$=14.74eV), for this pan of transitions the constant (C) can be calculated and is equal to 0.5 $eV^{-0.75}$.

Also the electron temperatures were measured in argon plasmas as a function of time, using the argon line emission intensity ratio of $4348A^0/4159A^0$ at (Ar^+) ($E_{i,g}$ =16.64+19.49=36.13eV), and $4159A^0$ (Ar^0) ($E_{P,g}$=14.47eV), for this pan of transitions we found that C is equal (0.8) $eV^{-0.75}$. In order to improve the accuracy Te was evaluated separately and all the temperatures were average. Figure (5) shows the electron temperature of Ar hollow cathode discharge variation during the first waveform of the discharge current at Ar pressure 10-2 torr, charging voltage 4kV.

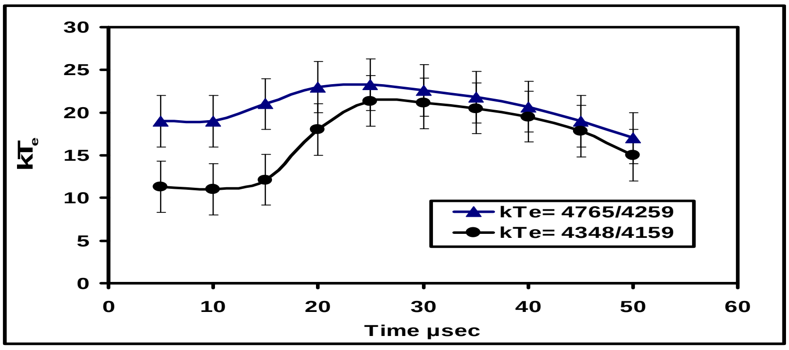

Fig. 5: the electron temperature of Ar hollow cathode discharge variation during the first wave of the discharge current at, charging voltage 4kV, at Ar pressure 10^{-2} torr

The increase in electron temperature with time in the range (5-25μsec.) is related to the increasing in the discharge current, more energetic secondary electrons are emitted from the surface of the hollow cathode which enhance the ionization events and as a results the electron temperature.

The addition of helium with small flow rate 5sccm to argon in the hollow cathode discharge can give information to estimate the electron temperature in the discharge using the ratio of the emitted line radiation of helium from the discharge. Helium has lower efficiency of cathode sputtering owing to its low mass compared to other inert gases (Wagatsuma, K. *et al*. 2001).

The second method to determine the electron temperature is using the intensity ratio of emission helium lines He $4713A^0/4921A^0$ and $5048A^0/4713A^0$. We used the sensitivity curve estimated by Easlund *et al*. 1973 to measure the electron temperature. Figure (6) show the electron temperature in argon discharge variation with time during the first period of discharge current at different gas flow rate (80-240sccm), charging voltage 4kv, 5sccm, helium gas.

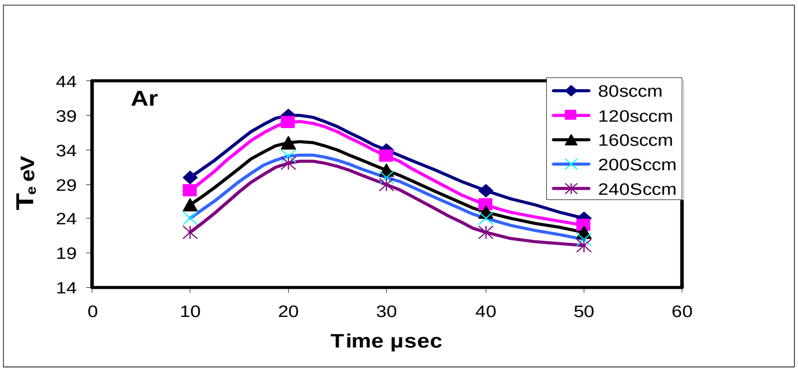

Fig. 6: the electron temperature of Argon hollow cathode discharge variation with time during the first period of discharge current with different gas flow rate (80-240Sccm), at charging voltage 4k.

Figure (6) shows that the electron temperature decreases with the increase in gas flow rate. The reduction of T_e with rise of gas flow rate may be explained as follows: when the gas flow rate increases, it causes an increase in the number of collisions between electrons and other plasma species. Consequently effective collisional transfer of energy occurs, that reduces the electron temperature.

Also gas flow rate increases further the plasma density is found to increase since the primary electron mean free path becomes smaller than the cathode diameter and gas ionization is less efficient and occurs closer to the cathode. As expected the electron temperature decreases slightly with increasing flow rate as thermal energy transfer to the neutrals increases.

The values of electron temperature measured by Helium impurity method are higher than that measured by argon line ratio. This is attributed that helium has the highest metastable energies among the other inert gases, making it a powerful Penning reagent for exciting plasma species through inelastic collisions (Sugimto, I. *et al*. 1994).

3.4.Measurements of Argon plasma density:

It was found that the ArII line intensities are proportional to the plasma density (Glen, P. Jacksona *et al*., 2003). For The ArII ($4880A^0$) line ($3P^4S – 3P^4(3P)4P$ [$2P – 2P_0$]. Since the ArII (ArII $4880A^0$) lower level is 17.14eV above the ArII ground state (Eexc, >> kTe) and the line intensity is proportional to the plasma density (n) (Qayyum, A. *et al*., 2005). Figure (7) shows the time resolved of intensity exited ion line ArII ($4348A^0$) and discharge current at charging voltage 4kV, Argon pressure 10^{-2} torr. During the high current discharge pulse a large amount of excited argon atoms, ArII ions and also long-lived atomic and ion metastables are produced. Two hump observed in the early afterglow of ArII lines indicate the existence of a complex mechanism of ArII energy levels population during the afterglow. The decrease of Ar emission is mainly due to reduced density of Ar atoms by local gas heating. This density decrease does not allow the emission from Ar atoms to reach its maximum simultaneously with that of current. The emission drop starts just with current growth due to reflection of energetic Ar atoms on the anode surface, resulting from acceleration of Ar ions towards the anode. The relative number of such atoms is small if the current is small, but grows considerably at high power discharge (Rossnagle, S.M. *et al*. 1988).

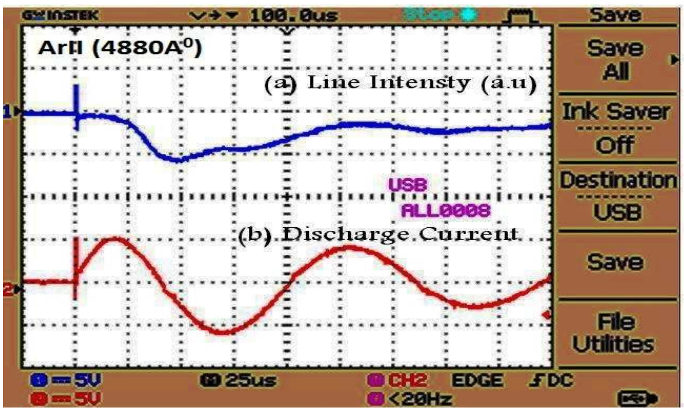

Fig. 7: Time resolved of intensity exited ion line ArII (4880A^0) and discharge current waveform at charging voltage 4kV, Argon pressure 10^{-2} torr.

The electron density depends on the neutral gas density. Since primary electrons are born at the cathode and scattered by inelastic and elastic neutral collisions, the primary electron density profile becomes hollow with increasing neutral density (Handle, F. *et al*. 1984). Figure (7) shows that the electron density decreases with time in the afterglow discharge resulting mainly from no more electron production by electron impact ionization because of the low electron temperature.

Conclusion:

DC pulsed cylindrical hollow cathode of charging voltage 4kV has been characterized by means of time resolved emission spectroscopy at different argon flow rates. The experimental results presented here suggest that electron temperature (T_e). T_e was determined using line to line intensity ratio method according to Corona model. Te increases with the discharge current, while it decreases with argon flow rate. The intensity variation of line emission of ArII (4880A^0) with time gives a true trend of the electron density. The electron density (n_e) decreases with time in the afterglow discharge because of recombination and diffusion processes. The present results are important for the usefulness of emission spectroscopy has a simple and reliable technique to measure the electron temperature without disturbing the plasma.

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