Effect of He and Ar Addition on N_2 Glow Discharge Characteristics and Plasma Diagnostics

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ABSTRACT

This paper reports the effect of inert gas mixing on the N_2 glow discharge characteristics and plasma parameters. The discharge current-voltage (I-V) characteristic curves of the discharge and Paschen's curves were measured at different gas mixture percentage. Langmuir probe is used for determination of electron energy distribution functions, electron temperature, plasma potential, ion density and electron density. The results demonstrate that the ampere-volt characteristics were different in (N_2-He) and (N_2-Ar) mixtures compared to pure nitrogen. The breakdown voltage increases by increasing He percentage in (N_2-He) gas mixture. Meanwhile it decreases by increasing Ar percentage in (N_2-Ar) gas mixture. The electron temperature and density can be either raised or reduced effectively by mixing helium or argon in a nitrogen discharge. The plasma potential follows the same trend as the electron temperature. The electron energy distribution function (EEDF) has two groups of electrons and can be either shifted effectively to low or high energy values by mixing helium or argon in a nitrogen discharge.

Keywords: Glow discharge/ plasma potential/ Paschen's curves/ Langmuir probes.

INTRODUCTION

Glow discharges are used in various applications such as deposition of thin films, etching and modification of surfaces in semiconductor industry and materials technology [1]. The low pressure glow discharge is a self-sustaining discharge with a cold cathode emitting electrons due to secondary emission mostly due to the ion bombardment. These secondary electrons lose their energy through inelastic and elastic collisions with the plasma species [2]. Addition of inert gases such as argon, neon and helium in nitrogen plasma, enhances the concentration of active species through penning excitation and ionization processes. The collision processes and plasma reactions in the mixture can be explained by knowing various plasma parameters such as electron temperature, electron density, plasma potential and electron energy distribution function (EEDF). It is well known that electron temperature can be tuned by adding inert gas in nitrogen plasma [3].

The advantage of Helium addition in nitrogen discharge over the other inert gases is that: it has lower efficiency of cathode sputtering owing to its low mass and thus can be added in nitrogen plasma for enhancing the production of active species without causing any considerable increase in the impurity level [3]. Also the addition of argon in nitrogen plasma makes the discharge stable and provides a better control over the plasma parameters [4].

In this work, the effect of inert gas mixing on the electrical characteristics and plasma parameters of DC N_2 glow discharge is investigated. In particular, argon and helium gases are mixed with nitrogen plasma in a plasma chamber at a constant operating pressure of 0.4 Torr. DC Langmuir probe is used to determine the plasma parameters (electron density and temperature, electron energy distribution function, ion density and plasma potential).

EXPERIMENTAL

Plasma Source

Figure (1) shows a schematic diagram of the experimental setup. A cylindrical discharge cell made of a Pyrex glass tube of 18 cm length and 13 cm diameter. Two parallel movable circular electrodes were enclosed in the discharge cell. The two electrodes were made of copper and each of them was 5 cm diameter. The discharge cell was evacuated to a base pressure down to 10^{-3} torr. The flow of nitrogen, argon and helium gases is monitored with mass flow meters whereas pressure in the chamber is recorded by using pirani gauge. The applied voltage was controlled by a DC power supply which can produce potential up to 2000 V.

Langmuir Probe Measurements

Langmuir probe is commonly used as low-temperature plasma diagnostic due to its easy and simple implementation compared to other plasma diagnostics [5]. From the probe data it is possible to determine a large variety of parameters characterizing the plasma including electron density, electron temperature and ion density. The method of the Langmuir probe measurements is based on its *I–V* characteristic the so-called probe characteristic. The Langmuir probe assembly consists of Tungsten wire of 0.30 mm diameter and 5 mm length, supported by a Pyrex glass tube. Probe current is measured for DC bias voltages ranging from - 100 to +100V.

The electron temperature, T_e, is determined in electron volt (eV) from the slope of the ln (*I*)–*V* curve of the probe in the transition region between floating potential and plasma potential by the following equation [6]:

$$T_e = e/(k_B \, dLn\,(I)/dV) \tag{1}$$

where e is the electronic charge, k_B is the Boltzmann constant, I is the electron current and V is the probe voltage.

The electron number density, n_e, in cm^{-3} is obtained from the electron saturation current (I_{Se}) using the following equation [6]:

$$n_e = \frac{4I_{Se}}{eA}\left[\frac{m_e}{2kT_e}\right]^{1/2} \tag{2}$$

where A is the probe area and m_e is the magnitude of electronic mass.

Assuming a Maxwellian energy distribution in the unperturbed plasma, the following simplified formula for a cylindrical prope may be used to determine the ion current in the OML regime [7]:

$$I_i = \sqrt{2}/pAen_i\,(|eV|/m_i)^{1/2} \tag{3}$$

where I_i is the ion current and m_i is ion mass. The slope of the linear relationship of the square of ion current I_i^2 versus prope voltage yields the plasma ion density, n_i, without knowledge of the electron temperature, in accordance with OML theory.

The electron energy distribution function, *EEDF*, are also important for understanding various plasma processes, such as electron impact ionization and excitation. In this study the EEDFs are calculated according to Druyvesteyn formula [8]:

$$F(e) = 2\left(2m_e\right)^{1/2}\left(e^3 A\right)^{-1} V^{1/2} \frac{d^2 I_p}{dV^2} \qquad (4)$$

where I_p is the probe current.

Another important plasma parameter is plasma potential, which plays a crucial role in controlling the energy of ions in material processing applications. In this study the plasma potential is found from the zero crossing point of the second derivative of the probe characteristics.

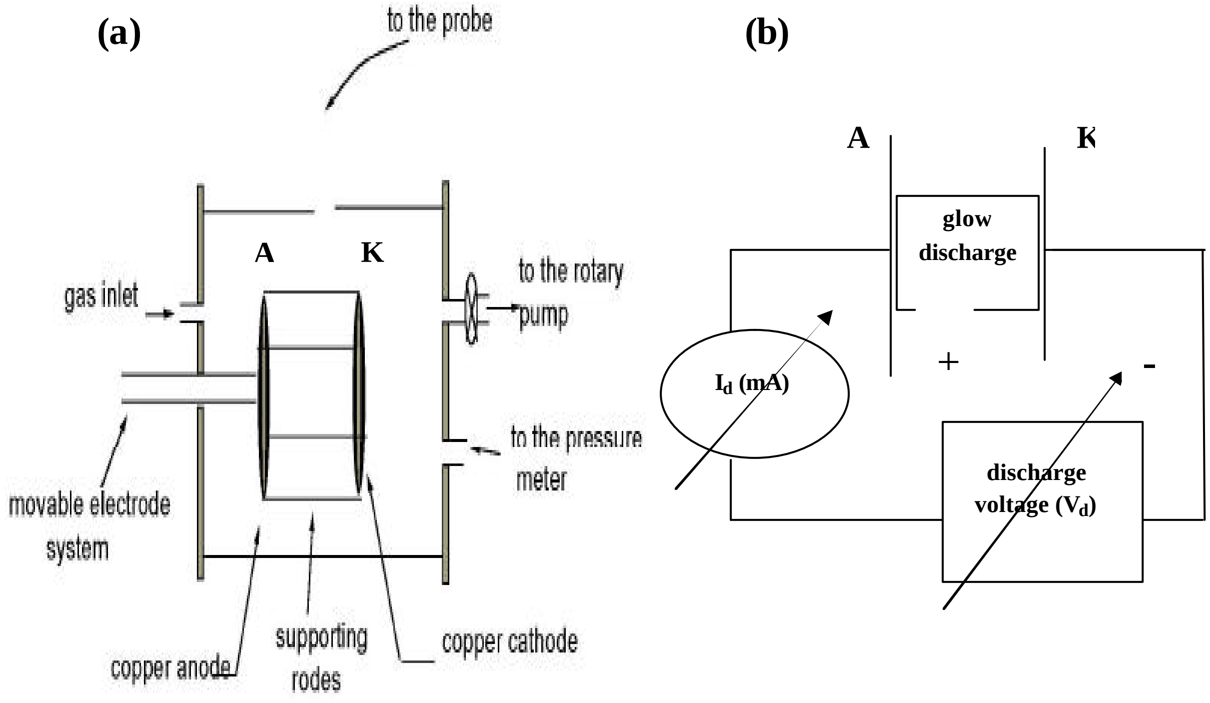

Figure (1): A schematic diagram for
(a) The experimental equipment and
(b) The electrical circuit

Current-volt

The (*I-V*) characteristic curves of N_2, at different He and Ar percentage in (N_2-He) and (N_2-Ar) mixtures were measured (Figure 2). The figure showed that the volt-amper characteristic were different in (N_2-He) and (N_2-Ar) mixtures compared to pure nitrogen discharges. More specifically, in (N_2-He) the discharge resistance increases yielding a lower electrical current at constant voltage and pressure, or a higher voltage at constant pressure and current. The decrease in the electrical current at constant voltage and pressure may be due to the following reason: the ionization cross-section of He is smaller than that of N_2 [3].

On the other hand in (N_2-Ar) mixture the situation is different. Its clear evidence that, the electrical current at constant voltage and pressure increases by increasing the Ar concentration from 0 up to 20%. With further increase of Ar percentage up to 40%, the electrical current decreases and then increases again up to 100% Ar. The increases of the electrical current by the increases of Ar percentage in (N_2-Ar) mixture can be explained as follow: the molecular and atomic forms of these gases have different excitation and ionization energies. Therefore, it is very often possible that the excited state of one form can ionize the other form (Penning effect) and causes the increase in the discharge current at the same voltage [9].

The decrease in the electrical current at constant voltage and pressure may be due to the following reason: the decreases of the Penning effect may be related to the increases of the number of argon atoms on the account of the nitrogen atoms [1].

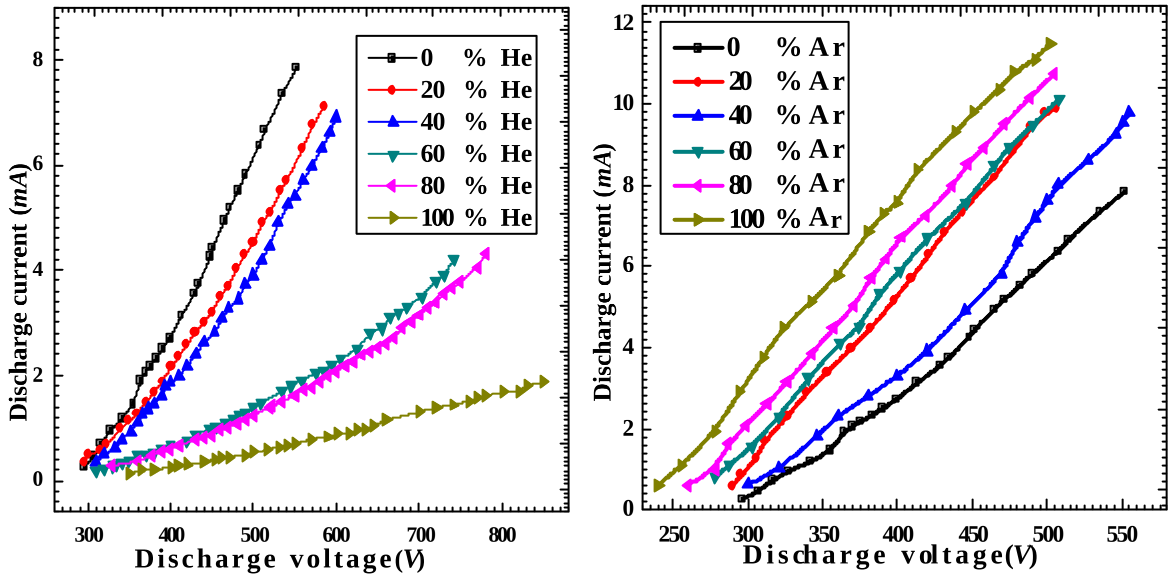

Figure : (2) The I-V characteristic curves of N_2, at different He and Ar percentage in the gas mixture and P = 0.4 torr.

Electrical breakdown and Paschen's curves

The glow discharges are widely applied for depositing thin polymer and oxide films, for cleaning the surface of materials, voltage stabilizers, etc. therefore the research into the conditions of the glow discharge breakdown is of considerable interest. As far as it is known, the breakdown curves of the glow discharge are described by Paschen's law, i.e. the breakdown voltage depends on the electrode spacing (d) and the gas pressure (P) as follows [10]:

$$V_b = \frac{BPd}{\ln(APd) - \ln[\ln(1+\gamma^{-1})]} \quad (5)$$

Where, *A* and *B* are constants found experimentally, which can be regarded as constants for a certain kind of gas, and γ is the secondary electron emission coefficient of the electrode. In the present work we investigate the influence of the gas composition on the breakdown voltage.

Paschen's curves were measured at different He and Ar percentage in the (N_2-He) and (N_2-Ar) gas mixtures and in the pressure range of 0.04 to 1 torr with the inter-electrode distance fixed at 7 cm (Figure 3). A general remark of these curves is that, all the curves have nearly the same general behavior and obey the standard curve. The breakdown voltage, V_b, initially decreases with increase in Pd, and then begins to increase with Pd after going through a minimum value [11]. On the left-hand side of the minimum Paschen's curves, V_b decreases fast when increasing Pd which can be attributed to the increase in collision frequency, equivalent to an increase in the number of collisions between electrons and neutral atoms or molecules. However, on the right-hand side of the minimum, V_b increases slowly with the increase of Pd, i.e. the ionization cross-section increases. Therefore, electrons need more energy to breakdown the discharge gap, resulting in an increase of V_b [12].

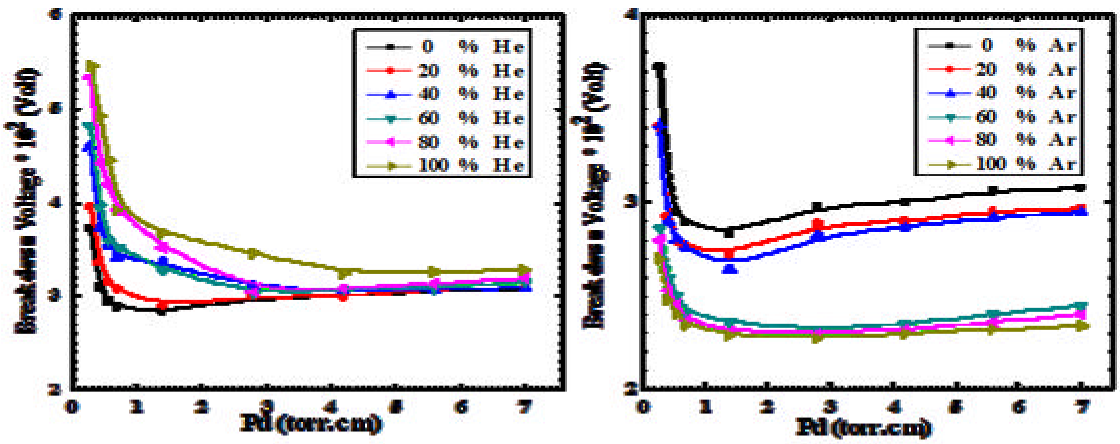

Figure: (3) Paschen's curves N_2, at different He and Ar percentages.

Also it has been found that the breakdown voltage increases by increasing He percentage in (N_2-He) gas mixture. This can be attributed to the higher ionization potential of helium as compared to nitrogen. On the other hand in (N_2-Ar) gas mixture, breakdown voltage decreases by increasing Ar percentage in (N_2-Ar) gas mixture. A final remark of these curves is that, the breakdown voltages in both (N_2-He) and (N_2-Ar) gas mixtures lie between those of the constituents. This result is similar to that reported by [13].

The electron temperature

Figure 4 shows the measured change of T_e, in nitrogen plasma when the concentration of argon and helium gas is varied. As the He concentration is increased, T_e increases. Meanwhile when argon is added to the nitrogen plasma, T_e decreases. This agrees with the results of papers [3, 14, and 15].

The increase of electron temperature in nitrogen plasma with the increase of helium percentage in the mixture may be explained as follows: helium has lesser electron collision cross-section than nitrogen, thus increase in the number of helium atoms in the mixture, decreases electron collision frequency and provides enough time for electrons to be accelerated owing to the electric field, which results as increase in kinetic energy of electrons [16]. On the other hand the decreases of electron temperature with argon addition may be explained as follows: the ionization cross sections and ionization potentials are nearly the same for Ar and N_2 [= 2.5 X 10^{-20} m^2 and =15.7 eV, respectively], so these parameters are not likely to explain the decrease in electron temperature with argon addition. The possible explanation is a difference in the EEDF especially in the high energy tail. When argon is added in the discharge, there is a sharp increase in electron density and hence a higher electron–electron collision frequency, which always tend to deplete electrons in the "hot tail" and the EEDF relaxes to Maxwellian. As a consequence, T_e decreases [14].

The electron density

Figure 4 shows the measured electron density as a function of mixing ratio in (N_2-He) and (N_2-Ar) discharges. For (N_2-He) discharges, the electron density decreases by increasing He percentage in (N_2-He) gas mixture. This result is similar to that reported in paper [14]. The electron density of (N_2-Ar) discharges increases with the mixing ratio. Also this result is similar to that reported in paper [14], even though a much smaller degree of density increase is found in our experiment. The electron density of (N_2-He) discharge is lower than that of (N_2-Ar) discharge because of the low ionization cross section of He [17].

The ion density

Figure 5 shows the measured ion density as a function of mixing ratio in (N_2-He) and (N_2-Ar) discharges. For (N_2-He) discharges, the ion density decreases by increasing He percentage in (He-N_2) gas mixture. This can be attributed to the low ionization cross section of He as compared to that of N_2. The ion density in (N_2-Ar) discharges increases with increasing argon content in the discharge and compared to the pure Ar discharge, the ion density in the pure N2 discharge is considerably lower under otherwise similar conditions. The ionization cross-sections and ionization potentials are nearly the same for Ar and N_2 [= 2.5 X 10^{-20} m^2 and = 15.7 eV, respectively], so that these parameters are not likely to explain the lower ion density in N_2 discharges. Thus a possible explanation is a difference in the electron energy distribution function in particular in the high-energy tail, which is indicated by detailed evaluation of the Langmuir probe characteristics. This is probably due to a higher secondary electron yield at the target in the Ar discharge [18].

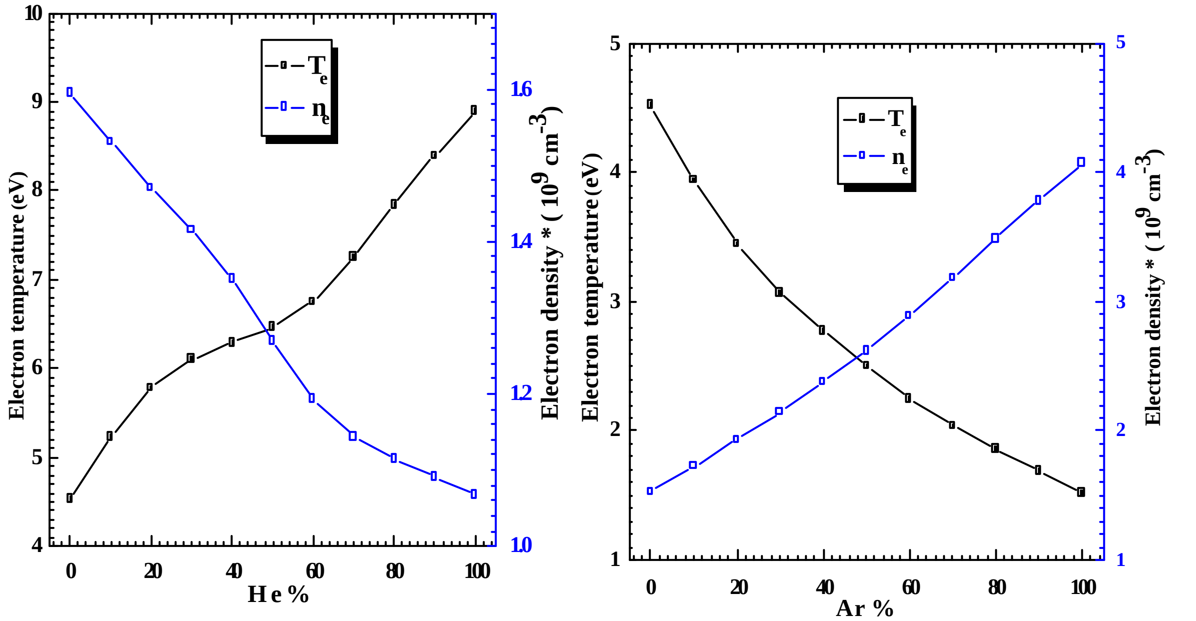

Figure: (4) Electron (temperature and density) variation with helium and Ar percentage in the mixture with 0.4 torr filling pressure at V_d = 500 V. discharge.

Electron energy distribution function

Figure 6 shows the measured electron energy distribution function (EEDFs) as a function of inert gas mixing ratio in nitrogen plasma. The plots indicate that (EEDFs) have two peaks. This represents two groups of electrons with different energies. The electrons in the first group, with low energy, affect local electron densities and the local plasma conductance. The electrons in the second group, with high energy, play the main part in the local excitation and the local ion production [19].

From the figure, it can be seen that there is a decided shift of the distributions to higher energy values as the helium percentage in the (N_2-He) mixture increased. The change in EEDF shapes with increase in helium percentage in mixture plasma is due to modification in various energy loss and gain reactions going on in the plasma. In addition to inelastic collisions of the electrons with molecules of nitrogen, there may exist superelastic collisions of electrons with metastable state of helium [3]. This will bring certain amount of low energy electrons to higher energy tail of the distribution function.

In case of (N_2-Ar) discharge, as argon percentage increases in the (N_2-Ar) mixture EEDF is shifted to low electron energy. This result is different from that reported by [20], where EEDF is shifted to high electron energy as Ar percentage increases in the mixture.

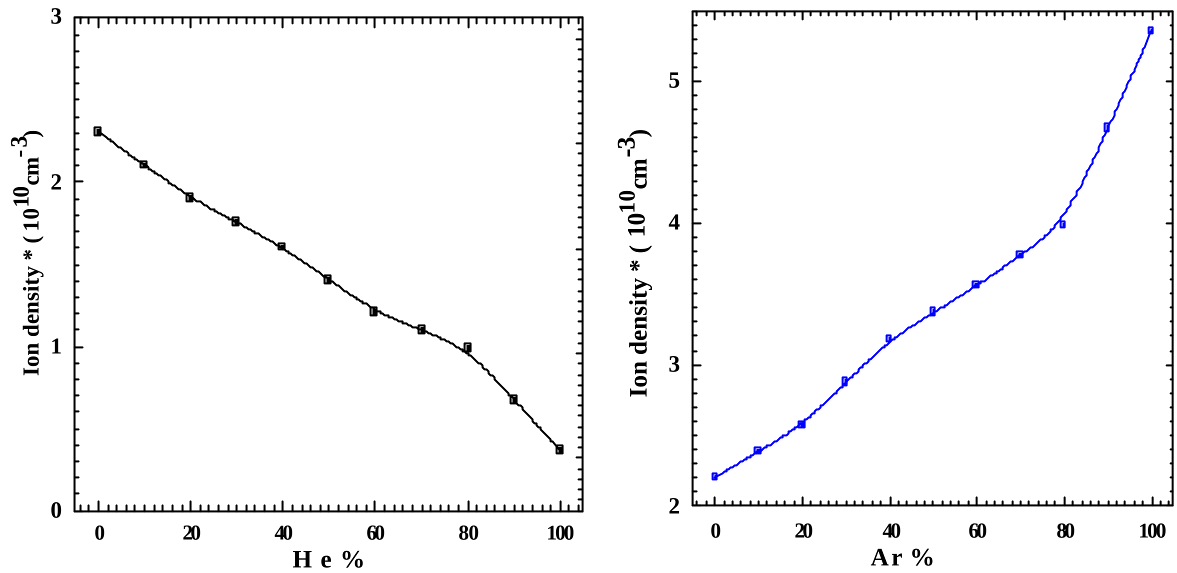

Figure (5): The ion density variation with helium and Ar percentage in the mixture with 0.4 torr filling pressure at V_d = 500 V.

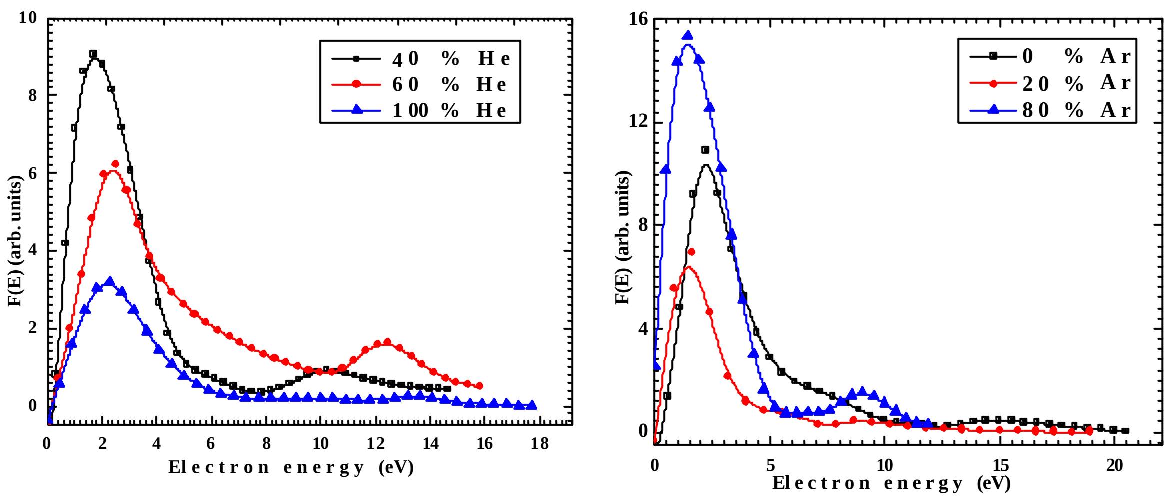

Figure (6): The electron energy distribution function at different He and Ar percentages, 0.4 torr filling pressure at V_d = 500 V.

Plasma potential

Figure 7 shows the plasma potential as a function of mixing ratio in (N_2-He) and (N_2-Ar) discharges. In (N_2-He) discharges, the plasma potential follows the same trend as the electron temperature (see Figure 4). This result is similar to that reported by [3]. A similar trend is also found in (N_2-Ar) discharges. This result is different from that reported by [21], where the plasma potential did not change with the mixing ratio.

The gradient of the plasma potential determines the electric field that is responsible for energizing the electrons, which maintain the discharge through ionization. Figure 8 represent a sample of the axial electric field distribution as a function of mixing ratio in (N_2-He) and (N_2-Ar) discharges. Due to the large potential drop in the cathode dark space, the axial field in this region is rather high and goes more or less linearly to virtually zero at the end of the cathode dark space region. So that it may be a good representative for the thickness of the discharge regions. The shapes of the electric field distributions do not significantly change with mixing ratio. The axial electric field distributions are quite similar for (N_2-He) and (N_2-Ar) discharges.

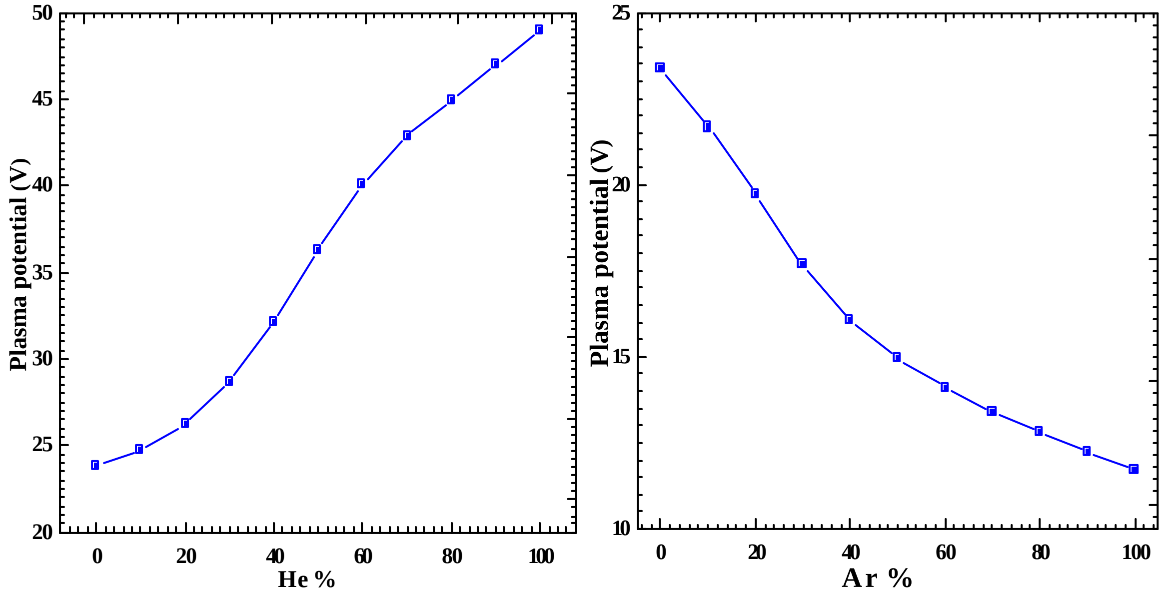

Figure (7): Plasma potential variation with He and Ar percentage in the mixture with 0.4 torr filling pressure at V_d = 500 V.

Figure (9) shows the cathode fall thickness, which measured from the potential distribution curves, as a function of mixing ratio in (N_2-He) and (N_2-Ar) discharges. In (N_2-He) discharges, the cathode fall thickness increases by increasing percentage of He in the gas mixture. Previous studies have shown similar results in DC (N_2- H_2) discharges using a single Langmuir probe [1]. In (N_2-Ar) discharges, the cathode fall thickness decreases by increasing percentage of Ar in the gas mixture.

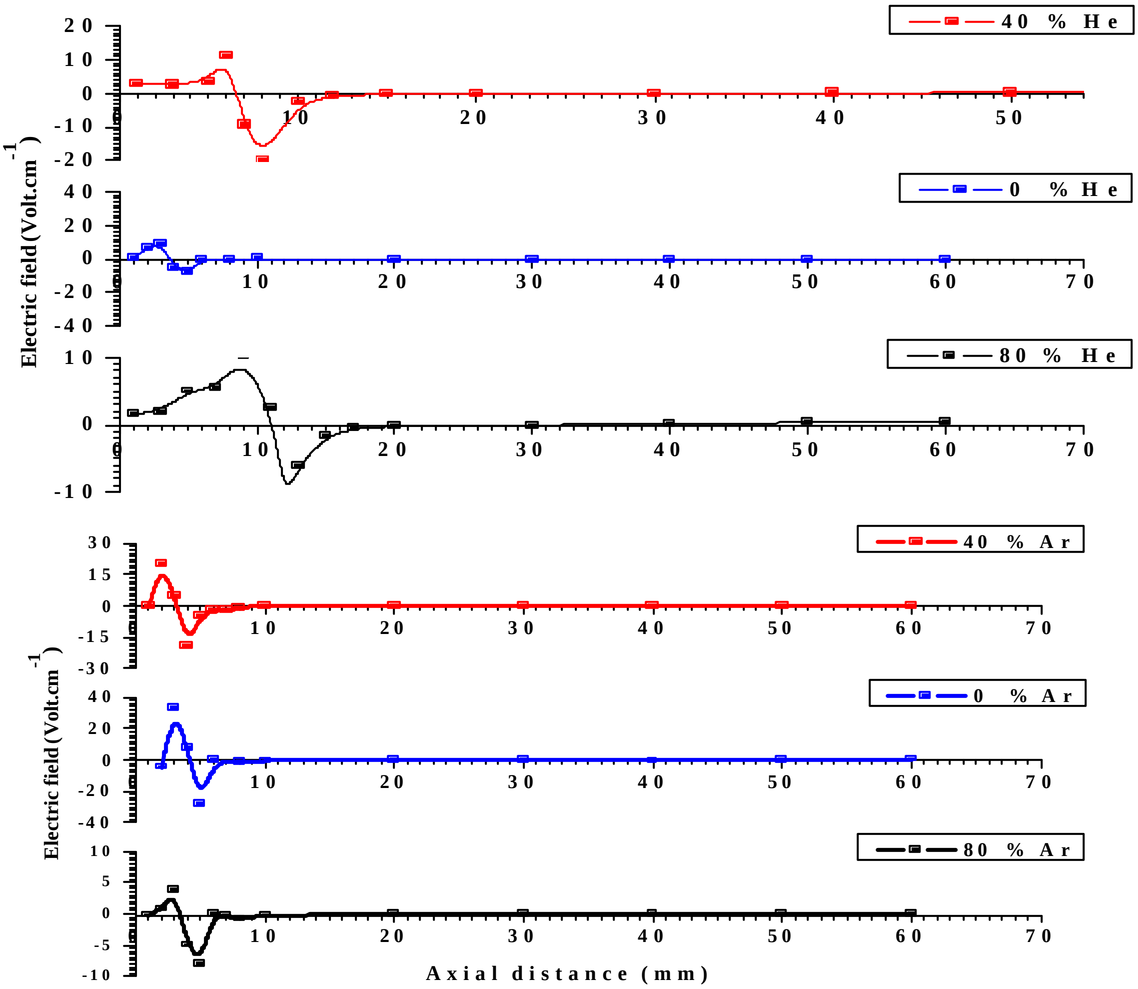

Figure (8): The electric field (E) as a function of distance from cathode at different He and Ar percentage in the mixture with 0.4 torr filling pressure at V_d = 500 V.

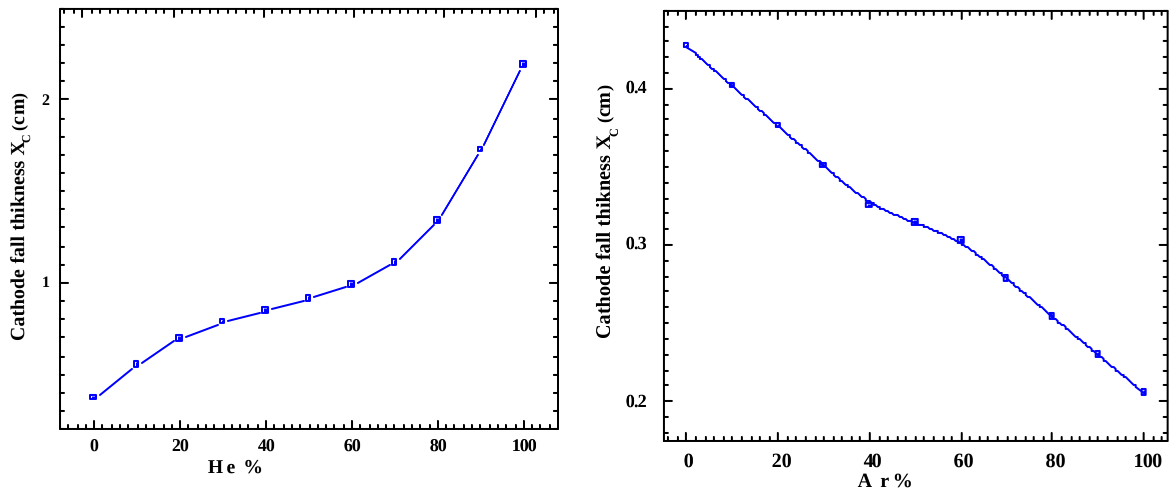

Figure (9): The cathode fall thickness (X_C) as a function of He and Ar percentage in the mixture with 0.4 torr filling pressure at V_d = 500 V.

CONCLUSION

I-V characteristic curves of N_2 discharge were studied at different helium and argon percentage in the discharge. Whenever the helium percentage was increased, the electrical current decreased under the investigated conditions. Meanwhile addition of argon from 0 up to 20% to (N_2-Ar) gas mixture resulting in increasing the electrical current. With further increase of Ar percentage up to 40%, the electrical current decreases and then increases again up to 100% Ar. The addition of argon or helium to N_2 discharge causes a dramatic change in the values of breakdown voltage of the discharge.

From single Langmuir probe measurements we can conclude that:

(1) As He concentration is increased in N_2 discharge, T_e increases. Meanwhile electron and ion densities decrease. When argon is added to N_2 discharge, T_e decreases while electron and ion densities increase.

(2) The EEDFs consist of two groups of electrons with different energies. There is a decided shift of the distributions to higher and lower energy values as the helium and argon percentage in the mixture increased.

(3) The shapes of the plasma potential and electric field distributions do not significantly change with mixing ratio.

(4)The cathode fall thickness increases/decreases by increasing percentage of He/Ar in the gas mixture.

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