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Full Length Research Paper

Electrical characteristics of (N_2-H_2) gas mixture DC glow discharge

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The electrical characteristics of DC Nitrogen-Hydrogen mixture glow discharge were investigated. The discharge current-voltage (I-V) characteristic curves of the discharge were measured at different gas pressures and gas mixture percentage. The axial distribution of both potential and electric field were studied at different H_2 percentage. The existence of the reversal electric field in the negative glow region was confirmed. Using the axial electric field distribution, the cathode fall thickness (X_c) was calculated. Second derivative of the single Langmuir probe electron current method was used to measure the electron energy distribution function (EEDF) in (N_2-H_2) gas mixture DC glow discharge at different H_2 percentage. In the positive column region, a Maxwellian EEDF was found at different conditions, while in both cathode fall and negative glow regions, a non-Maxwellian EEDF was observed. Two groups of electrons were detected in these two regions.

Key words: Glow discharge, axial potential and electric field distributions, 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. The low pressure glow discharge plasma is created by applying a potential difference between two electrodes of certain distance apart. Since there is a continuous loss of electrons in the discharge, there must be an equal degree of simultaneous ionization to maintain a stable discharge. Additional electrons are produced by secondary emission from the cathode surface (Grill, 1993).

A glow discharge is characterized by its distinctive regions between the cathode and the anode electrodes across the tube. The physical characteristics of these regions depend on the parameters of the discharge that is, geometry of the discharge tube, pressure of the working gas, type of gas, cathode material, applied potential and the discharge current.

The DC glow discharge in different Ar-N_2 and H_2-N_2 gas mixtures results in the surface nitriding of titanium metal and its alloy. Various gas mixtures were used in order to establish the main active species governing the nitriding process that is N, N_2 or NH as excited or ionized particles. The results showed enhancement of the hardness of the treated specimens and that the glow discharge gas pressure critically influences the nitriding process.

Some of the advantages of the nitriding using a DC glow discharge include rapid kinetics of reaction at the surface, homogeneity of the treated surfaces, good control and intrinsic non-toxicity of the process.

Rusnak and Vicek (1993) used emission spectroscopy as a plasma analysis technique in the cathode fall and the negative glow of (N_2-H_2) abnormal glow discharge for steel surface nitriding. They found that the addition of small amount of hydrogen leads to significant increase in the discharge current and, as a consequence, in the cathode surface temperature. They studied the effect of different percentages of (N_2-H_2) gas mixture on the electrical characteristics of the discharge such as the *I-V* characteristic curves, the breakdown voltage, the cathode fall thickness, the axial distribution of potential and electric field.

Wei et al. (2007) studied the influence of H_2 addition on the discharge characteristics of (Ne-Xe) in alternating current plasma display panel. The operation voltage margin affects both the H_2 % and the sustaining voltage fre-

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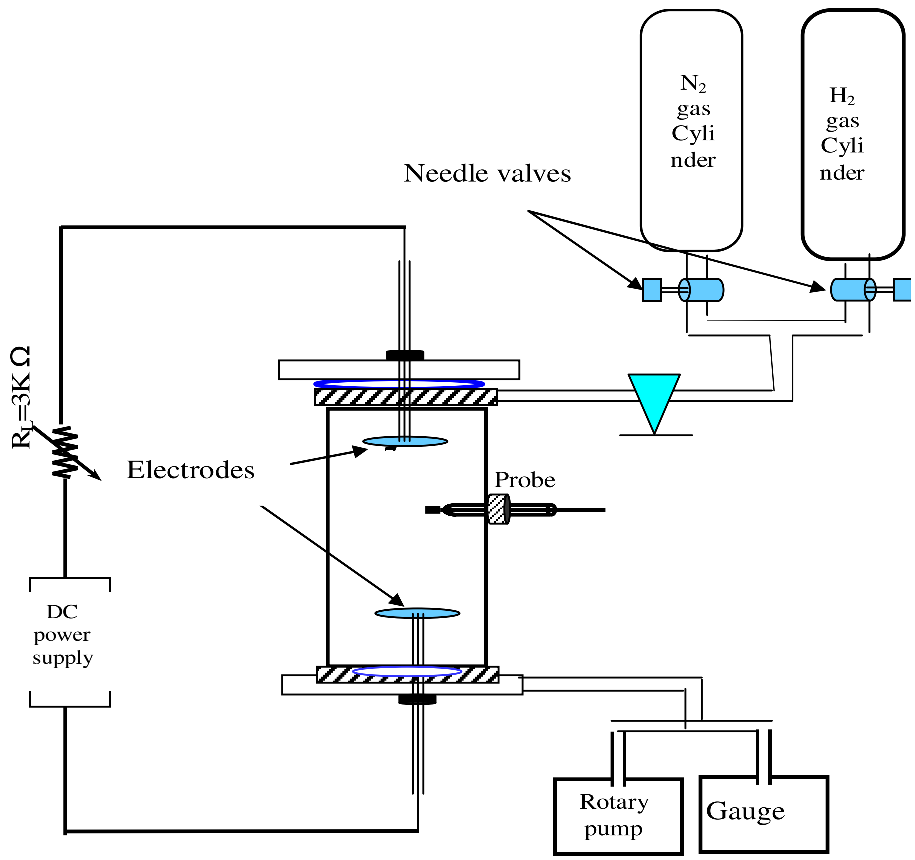

Figure 1. A schematic diagram of the experimental setup.

quency. The operation voltage margin increases firstly and then decreases with increasing H_2 %. Also, the maximum value of operation voltage margin can be obtained at a certain H_2 %. The discharge current increases remarkably when a little H_2 is added to the (Ne-Xe) gas mixture discharge.

Menéndez et al. (2005) studied the addition of hydrogen to a direct current (DC) argon glow discharge (GD) coupled to a time of flight mass spectrometer using a fixed voltage between the electrodes and a fixed discharge pressure. Results have shown that hydrogen addition gives rise to significant changes in the slope of the linear relationship between the electrical current and the discharge voltage. Clearly, the electrical resistance of the discharge at the typical operation voltages in the interval 600 - 1000 V increases with hydrogen added to pure argon. A decrease of the sputtering rates was observed and this results in a higher hydrogen concentrations. Wang et al. (2009) studied the effects of DC plasma nitriding parameters on the structural phases, micro-hardness and dry-sliding wear behavior of the nitrided layer were investigated by optical microscopy, X-ray diffraction, and scanning electron microscopy, micro-hardness testing and ring-on-block wear testing.

Hosseini et al. (2009) measured Nitrogen depth profile of plasma nitrided pure iron and evaluated by accurate experimental techniques. Plasma nitriding cycles were carried out on high purity iron substrate in an atmosphere of 75% H2, 25% N2. Nitrogen concentration depth profiles in the compound layer and the diffusion zone were characterized by glow discharge optical emission spectroscopy (GDOES) and secondary ion mass spectroscopy (SIMS), respectively. Nitrogen diffusion depths were measured accurately by optical and scanning electron microscopy as well as SIMS technique at different nitriding times. Experimental results indicated good agreement between SIMS data and microscopic evaluations for various nitriding cycles.

The aim of the present work was to study the effect of the changes in H_2 percentage on the electrical characteristics of (N_2-H_2) mixture glow discharge and also, determine EEDF using single Langmuir probe technique at different discharge conditions and at different H_2 percentage in (N_2-H_2) gas mixture.

THE EXPERIMENTAL SETUP

Figure (1) shows a schematic diagram of the experimental setup. A cylindrical discharge cell made of a Pyrex glass tube of 23 cm length and 7 cm diameter. Two parallel movable circular electrodes

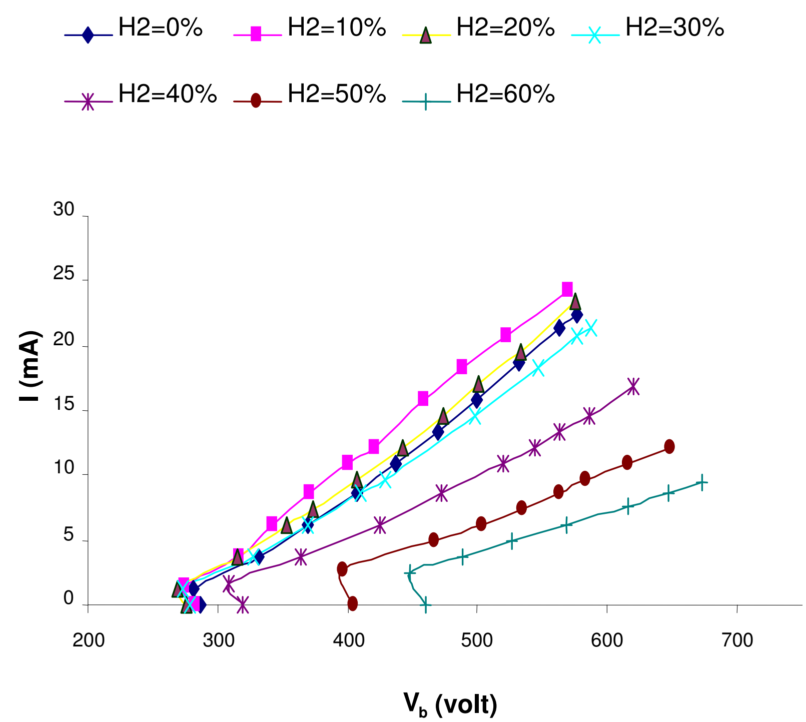

Figure 2. The I-V characteristic curves at different H_2 percentage in the gas mixture at separation distance (d) = 1.5 cm and p = 1 torr.

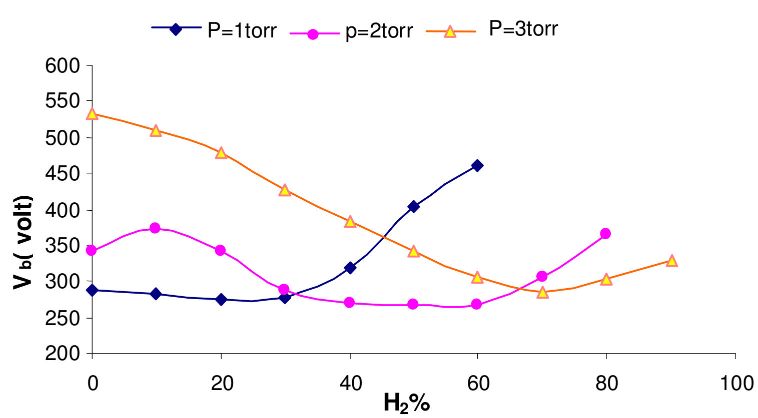

Figure 3. The breakdown voltage (V_b) as a function of H_2 percentage at d = 1.5 cm.

were enclosed in the discharge cell. The two electrodes were made of stainless steel and each of them was 5 cm diameter. The discharge cell was evacuated using two stage rotary pump (type Tungsram) to a base pressure of 7 motor and the gas was then admitted to fill the tube at the desired flow rate and gas pressure. A vacuum gauge (type Edwards capsule dial gauge cg 16 K) was connected to the discharge tube to measure the pressure of the gas inside it. The cell was filled with the working gas (hydrogen and nitrogen gases). Two needle valves were used to control the flow rate of the gas mixture inside the cell.

The applied voltage was controlled by a DC power supply which can produce potential up to 1000 V and current up to 100 mA.

Single Langmuir probe, made of molybdenum wire (of 0.3 mm diameter) was used. The wire is isolated by a thin glass tube. A tip of 2 mm length of the wire is immersed in the discharge tube.

EXPERIMENTAL RESULTS AND DISCUSSION

The I-V characteristic curves of the discharge

The I-V characteristic curves, at different gas pressures and at different H_2 percentage in (N_2-H_2) gas mixture were measured (Figure 2).

Figure 3 represents the breakdown voltage (V_b) as a

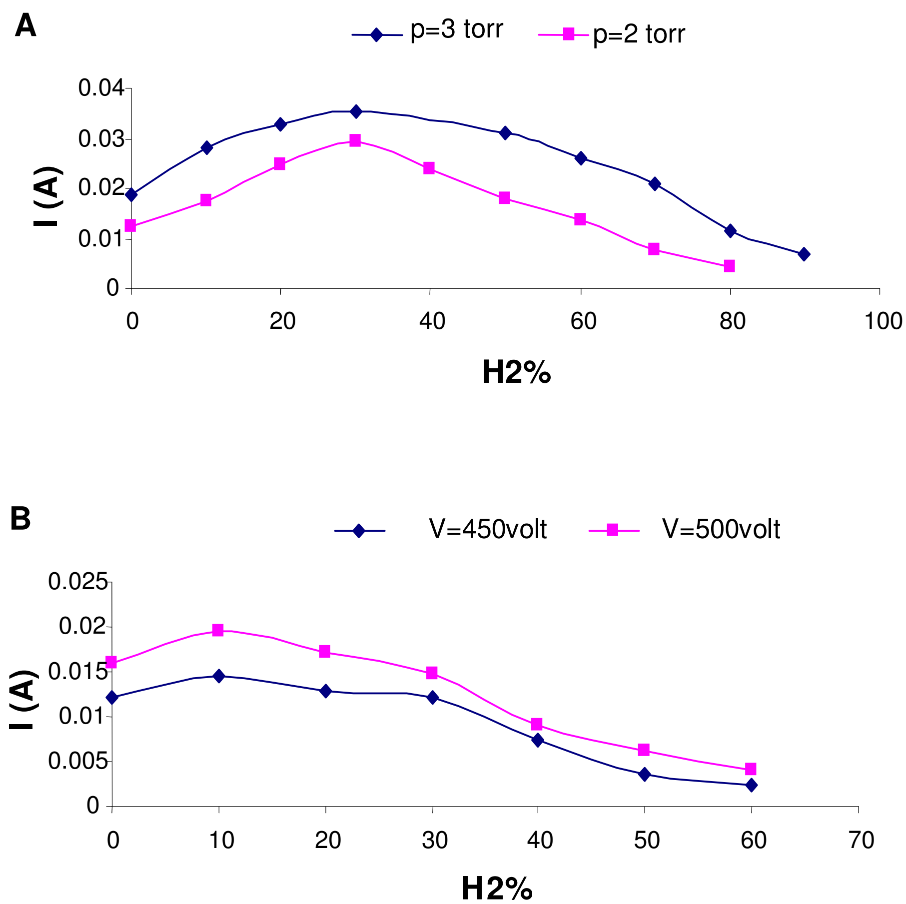

Figure 4. The discharge current (I) as a function of H_2 percentage at p = 1 torr: (a) at different gas pressure; (b) at different discharge voltage.

function of H_2 percentage in (N_2-H_2) the gas mixture at different gas pressures. Its clear evidence that, the breakdown voltage V_b, at a constant separation distance between the two electrodes (d = 1.5 cm) increases by increasing the gas pressure for H_2 concentrations from 0 up to 40% which follow the right hand side of Paschen's curve. With further increase of H_2 percentage, the breakdown voltage V_b decreases by increasing the gas pressure which follows the left hand side of Paschen's curve. This means that, the change of H_2 percentage in (N_2-H_2) gas mixture cause a wide change in the gas characteristic. The breakdown voltage decreases by an additional increase in H_2 percentage of the mixture until a certain value, which depends upon the total pressure of the gas, which then leads to a further increase in the breakdown voltage.

On the other hand, Figure 4 shows that at the same applied voltage, the discharge current increases by an additional increase in the H_2 percentage and then decreases again.

The increases of the discharge current by the increases of H_2 percentage in (N_2-H_2) gas mixture can be explained as follow: The increase of secondary electron emission coefficient due to reduction of the surface oxides and surface cleaning by hydrogen as mentioned by Rusnak and Vicek (1993) and Hulett and Taylor (1985).

Also, 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 (Nasser, 1971).

The decrease in the discharge current at the same applied voltage may be due to the following reasons:

1). The decreases of the Penning effect may be related to the increases of the number of hydrogen atoms on the account of the nitrogen atoms.

2) The ionization cross-section of H_2 is smaller than that of N_2 (Von Engel, 1983).

Paschen's curves

Paschen's curves were measured at different electrode separation distances (d) and different H_2 percentage in the (N_2-H_2) gas mixture.

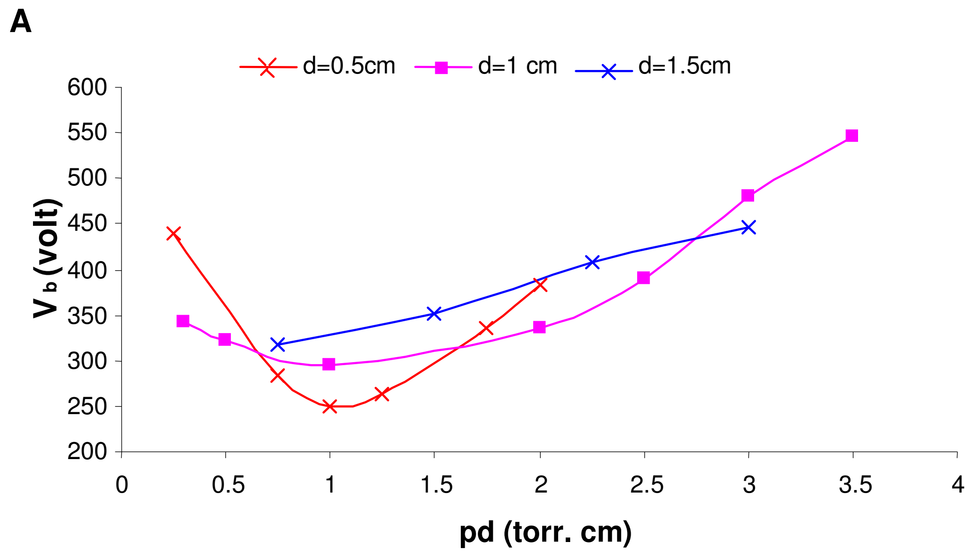

Figure 5a. Paschen's curves at different separation distances.

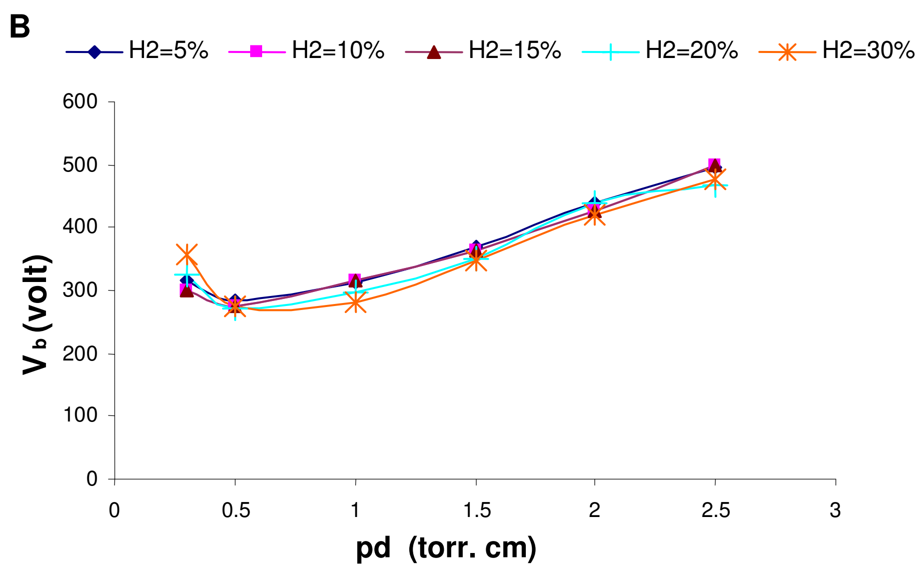

Figure 5b. Paschen's curves at different H_2 percentages.

Figure 5a shows the Paschen's curves at distance d = 0.5, 1, and 1.5 cm for H_2 = 20 %. From these curves, the minimum breakdown voltage ($V_{b\ min}$) occurs at pd_{min} of 1 torr. cm at the electrode separation distance of 0.5 cm. When the separation distance increases than 0.5 cm, the Paschen's curves deviate from the standard curves. This deviation was also noted by Garamoon et al. (2003). They explain the deviation on the basis of the change in the active secondary processes. The numbers of photons and exited atoms which are produced near the anode and reach the cathode depend on the solid angle between its point of product and the cathode. If the ratio of the electrode diameter to the inter electrode distance is small, then the solid angle will be small and few photons and excited atoms will reach the cathode. On the other hand, Figure 5b shows the Paschen's curves at different H_2 percentage in the (N_2-H_2) gas mixture at p = 0.5 torr.

Axial distribution of potential and electric field

Figure 6 shows the potential distribution V at different H_2 percentage. The potential increases rapidly near the cathode until it reaches a maximum value at the end of cathode fall region, this rapid increase in V can be referred to the existence of positive space charge within the

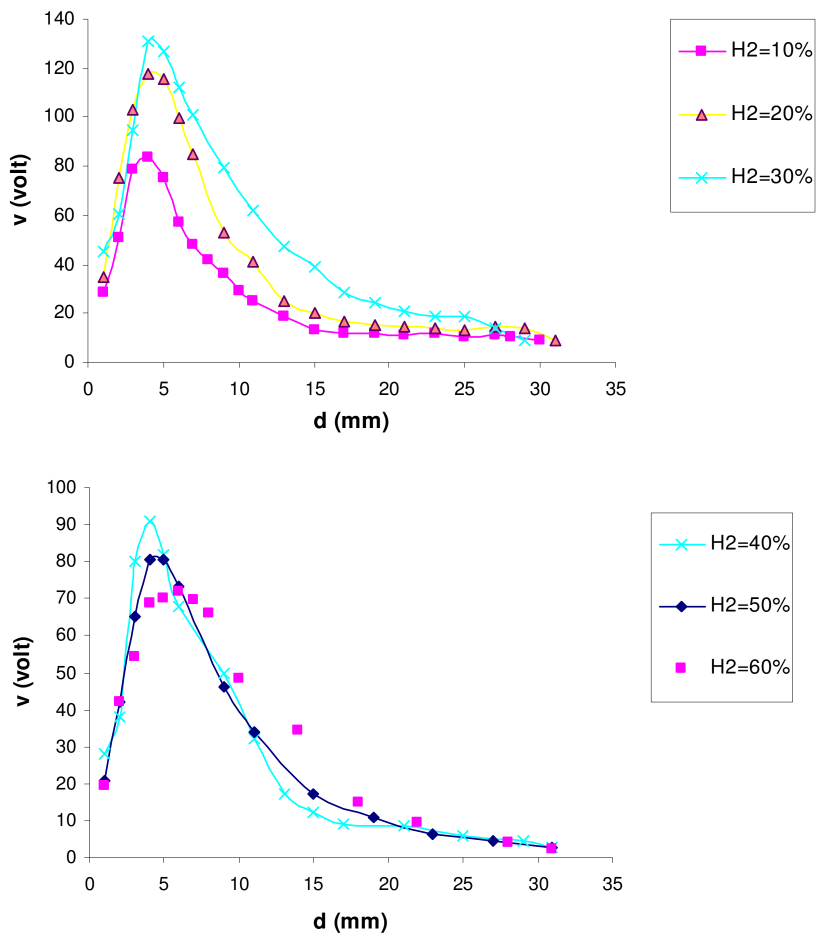

Figure 6. The potential distribution as a function of the separation distance from the cathode at p = 1 torr for different H_2 percentage.

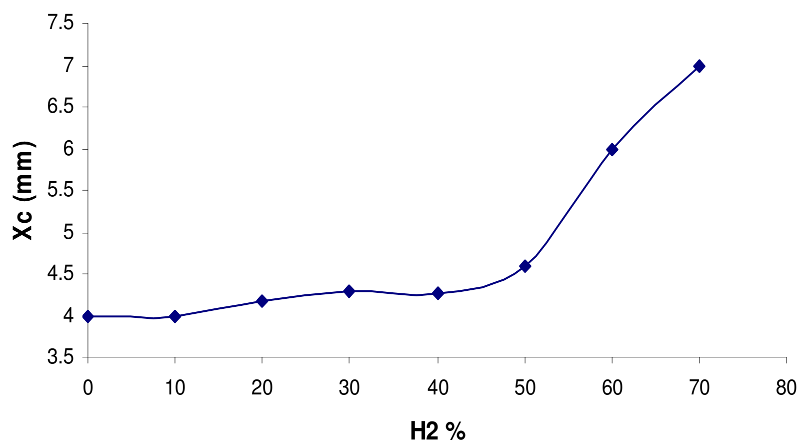

Figure 7. The cathode fall thickness (X_c) as a function of H_2 percentage.

cathode fall region. Then, the potential decreases less rapidly due to the existence of the reversal field in the negative glow and at the beginning of Faraday dark space.

Figure 7 shows the cathode fall thickness, which measured from the potential distribution curves, as a function of H_2 percentage in the (N_2-H_2) gas mixture. The cathode fall thickness (X_c) increases by increasing percentage of

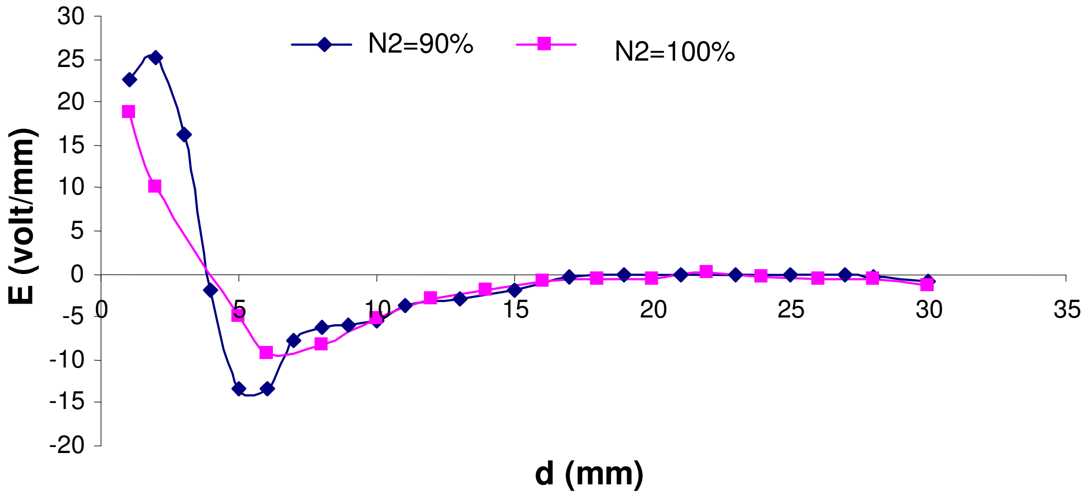

Figure 8. The electric field (E) as a function of distance from cathode at different N_2 percentage.

H_2 in the gas mixture (Rusnak and Vicek, 1993).

The electric field is determined by differentiating the applied potential (V) with respect to the distance between the two electrodes (X). Figure 8 shows values of the electric field distribution at different H_2 percentage in (N_2-H_2) gas mixture. The electric field has large values near the cathode electrode and decreases sharply until its reaches zero at the end of the cathode fall and reverse its direction again. The reversal of the electric field can clearly be observed within the abnormal glow operating conditions. At a certain position in the negative glow, the electric field changes its sign and tends to confine the electrons in the negative glow, meanwhile, driving some of the ions towards the anode. The position of the field reversal depends only on the electrode separation distance, the length of the cathode sheath and the energy relaxation length of the fast electrons (Maric et al., 2002)

Figure 9 shows the reverse electric field thickness, X_r, which was inversely proportional to the gas pressure. The data clearly show the shift of the field reversal towards the anode as the gas pressure decreases.

Measurements of electron energy distribution function

Electron energy distribution function (EEDF) was determined in the present work at different H_2 percentage in the (N_2-H_2) gas mixture using single probe technique. EEDF was determined using the second derivative method of the electron current of the I-V characteristic curves of the single probe (Swift and Schwar, 1970).

$$F(\varepsilon) = const\sqrt{V_p}\exp\left(-\frac{eV_p}{kT_e}\right)$$

For Maxwellian energy distribution

$$\frac{dI^2}{dV_p^{\ 2}}\alpha\exp\left(\frac{-eV_p}{kT_e}\right)$$

Thus;

$$F(\varepsilon) = \frac{4}{A_p e^2}\left(\frac{m_e V_p}{2e}\right)^{\frac{1}{2}}\frac{d^2 I}{dV_p^{\ 2}}$$

Where;

Ap is the probe area, ε = the electron energy and $\frac{d^2I}{dV^2}$ = the second derivative of the probe electron current with respect to the probe voltage.

Typical curves of this method were shown in Figure 10 which show the EEDE in the positive column region of the glow discharge at different H_2 percentage and at p = 3 torr. The results in this figure show that only one group of electrons appears and the distribution may be Maxwellian distribution.

On the other hand, two peaks were found in Figure 11, in the negative glow and faraday dark space regions of the glow discharge. This represents two groups of electrons with different energies. The reason for this effect was the beam of the primary electrons with high energy, which gain energy from the strong electric field near the cathode. So these electrons represent the high energy group moving from the cathode dark space region toward the anode region. In the negative glow region, they excite and ionize gas atoms without significantly changing their direction. The secondary electrons created

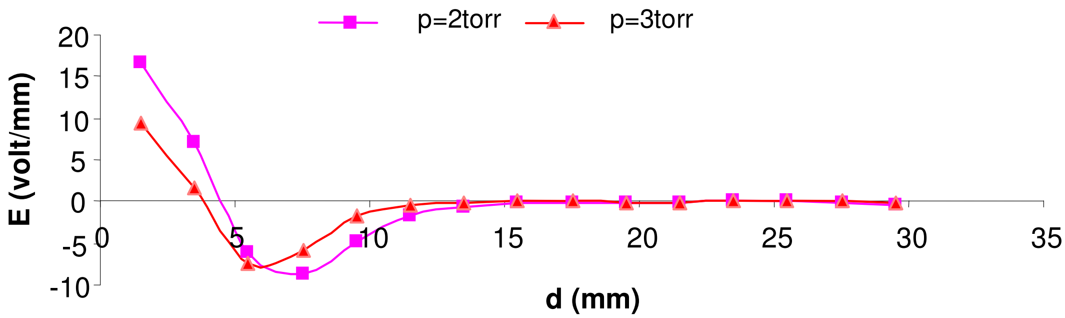

Figure 9. The electric field as a function of distance from the cathode at different gas pressure.

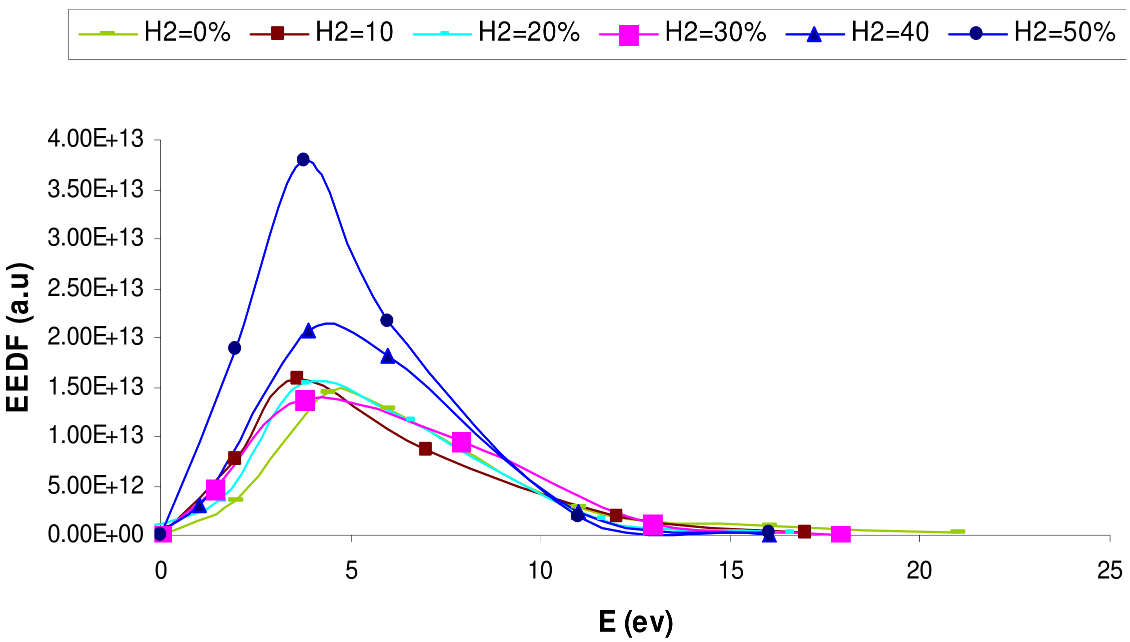

Figure 10. The electron energy distribution function in the positive column region as a function of H_2 percentage and at I = 20 mA and p = 3 torr.

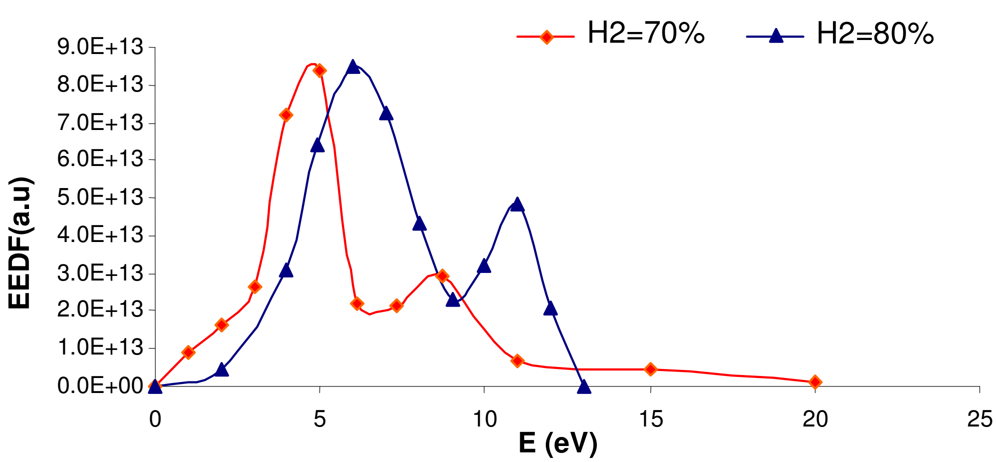

Figure 11. The electron energy distribution function in the negative glow region at I =20 mA and p = 2 torr.

electrons created by ionization also excite and/or ionize neutral atoms and thus form a group of low energy electrons. The low energy electrons and the secondary electrons diffuse towards the anode region (Popov et al., 2004). Meanwhile, because the long path causes the electron groups to have a longer time to redistribute (that is to thermalize) themselves, forming one group with Maxwillian distribution in the positive column region.

Elakshar et al. (2000) measured the electron energy distribution functions in the cathode fall and negative glow regions of the Ar and He glow discharge using a single Langmuir probe. Two groups of electrons having different temperatures were detected in these two regions for both gases. The presence of these two groups was attributed to a “runaway" process in the weakly ionized gas in which the conditions for electron acceleration without inelastic collisions were well satisfied. The electrons collide with neutral atoms in the negative glow region and produce secondary electrons of different temperatures.

Conclusion

I-V characteristic curves of the glow discharge were studied at different hydrogen percentage in a gas mixture of (H_2-N_2). Whenever the hydrogen percentage was increased upon a certain value, the discharge current increased. Further increasing of the hydrogen causes a decrease in the discharge current.

Paschen's curves were determined at different separation distances and pressures at different hydrogen percentage. At the same pd, the breakdown voltage (V_b) was increased by increasing the separation distance.

Using the numerical differentiation of the potential with respect to the distance from the cathode, axial distribution of the electric field was obtained at different gas pressures and hydrogen percentage. The cathode fall thickness (X_c) was estimated using axial electric field distribution and it was in the range of 0.5 cm.

Existence of the reversal electric field in the negative glow region of the glow discharge was confirmed experimentally. The reverse field thickness decreased by increasing the gas pressure.

Using Langmuir single probe, the electron energy distribution function (EEDF) was measured at different hydrogen percentage. It is concluded that, the EEDF in the positive column may be Maxwellian at different conditions. While EEDF in both cathode fall and negative glow regions showed that there are two groups of electrons and this became a non Maxwellian distribution.

REFERENCES

Elakshar FF, Hassouba MA, Garamoon AA, Fizika A (2000). Measurements of the electron energy distribution function in two different regions of DC-magnetron sputtering device pp.9-177.

Garamoon AA, Samir A, Elakshar FF, Kotop EF (2003). Plasma Sources Sci. Technol. pp.12-417.

Grill A (1993). "Cold Plasma in Material Fabrication from Fundamental to Applications ", IEEE Press.

Hosseini SR, Ashrafizadeh F (2009). Vacuum 83: 1174.

Hulett DM, Taylor MA (1985). Metal Progress pp.18-21.

Maric D, Kutasi K, Malovic G, Donko Z, Lj Z (2002). Petrovic, Eur. Phys. J, Appl. Phys. pp.21-73.

Menéndez A, Rosario P, Nerea B, Alfredo S (2005). Spectrochimica Acta Part B: Atomic Spectroscopy pp.60-824.

Nasser E (1971). "Fundamental of Gaseous Ionization and Plasma Electronics ", (Wiley-Interscience, New York.

Popov Tsv K, Dimitrova M, Dias FM (2004). Vacuum pp.76-417.

Runsak K, Vicek J (1993). Emission spectroscopy of the plasma in the cathode region of N2-H2 abnormal glow discharges for steel surface nitriding, J. Phys. D: Appl. Phys. 26: 585-589.

Swift JD, Schwar MJR (1970). “Electrical probes for plasma diagnostics”, London ILiffe book Ltd.

Von Engel VA (1983). "Electric Plasmas their Nature and Uses ", Taylor & Francis Ltd, London and New York.

Wang J, Xiong J, Peng Q, Fan H, Wang Y, Li G, Shen B (2009). http://www.sciencedirect.com/science?_ob=ArticleURL&_udi=B6TXJ-4TCR1RH1&_user=10&_coverDate=03%2F31%2F2009&_alid=1007629248&_rdoc=39&_fmt=high&_orig=mlkt&_cdi=5592&_sort=v&_st=17&_docanchor=&view=c&_ct=1715&_acct=C000050221&_version=1&_urlVersion=0&_userid=10&md5=4f024da706f39ff9a1c9e50c7f7e2a02 Mater. Characterization 60: 197.

Wei W, Jian S, Bingang G (2007). The influence of H_2 addition on the discharge characteristics of Ne-Xe in color alternate current plasma display panel". Eur. Phys. J. Appl. Phys. pp.37-331.