GAS TYPE ROLE ON THE DYNAMICS OF CHANNEL SPARK PULSED ELECTRON DEPOSITION SYSTEM

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ABSTRACT

In this paper, the effect of feeding gas type on the dynamics of channel spark pulsed electron deposition system is investigated. Electrical, magnetic, and optical characterisations of the system were measured for different feeding gases, oxygen (O₂), nitrogen (N₂), and argon (Ar). The discharge current for each gas type was measured with maximum value of 1189 A for O₂ at -13 kV applied voltage. The discharge current and voltage waveforms were simulated by LRC circuit theory. Effect of gas pressure on the maximum discharge current and total inductance was also investigated. The beam current investigated by faraday cup and reached maximum electron beam current of 136 A for O₂ gas. Two magnetic pickup coils were employed for the measurements of beam kinetic dynamics and the measured beam speed was around 0.4×10⁶ m/sec. Electron beam plasma density was calculated from faraday cup and magnetic coils signals and found to be 1.96×10²⁰ m⁻³. Optical emission spectra were also measured to identify reactive species and its role in electron beam interaction with graphite target for thin film deposition application. The ability of using the system for thin film deposition is demonstrated by depositing amorphous hydrogenated carbon (a-C:H) films over silicon substrates.

Keywords: Channel spark, Pulsed electron deposition, Thin film deposition, Amorphous hydrogenated carbon.

1. INTRODUCTION

Pulsed deposition devices allow exceptional material deposition technology. The short pulse length, on the order of about 10-100 ns, results in high power density at the target material surface. This leads to generation of rapid heating process and formation of a plasma plume. On the other hand, commercial production of pulsed deposition devices on large scale is limited due to high costs which decrease the efficiency of such techniques. Thus, it is essential to find a technology permitting both high throughput deposition characteristics and low running costs. These criteria can be realised by pulsed electron deposition (PED). Channel spark pulsed electron beam system is a promising technique for thin film deposition. A comparative study among pulsed intense electron beam systems showed that, Channel spark system has higher beam intensities and longer pulse lengths at low voltages than other systems [1-3].

The electron beam generated in the channel spark device is the most suitable pulsed electron beam source for material processing. Channel spark is a kind of pulsed electron deposition (PED) devices that recently has attracted considerations in many fields of materials processing. This is mainly due to the higher energy transfer to the target material as well as low running costs when compared to pulsed laser deposition (PLD) [4]. Ideally, pulsed electron beams are generated in transient hollow cathode discharges employing low gas pressures (10⁻³–10⁻¹ mbar) and discharge voltage on the order of few kV up to tens of kV [5,6].
Afterwards, a novel PED system called Channel Spark was designed allowing higher conversion efficiency of electrical energy stored. This conversion efficiency reached 30% compared to only 4% of pseudo spark discharge. Despite its simplicity, only with a suitable device design, one can attain beam parameters suitable for PED (e.g., electron beam current, energy, and plasma density) [7,8].

Practically, the focused electron beam extracted by the plasma passes through a tiny cathode hole and is guided into a 4-6 mm diameter tube to the target material. Normally this tube is made of glass or alumina or any other dielectric material. The high potential difference applied between the hollow cathode and the target holder (ground) results in an acceleration of the electron beam. To store the electrical energy high voltage ceramic capacitors are employed. These capacitors can deliver an energy of about 3-5 J/pulse. An external triggering circuit is used to initiate the discharge. Because of the focusing of the beam (several mm² in area), high current density in the beam can be achieved (about 10⁶ A/cm²). This leads to very high-power density (up to 10⁹ W/cm²) onto the surface of the target material. Subsequently, the energy is absorbed by the surface and a fast rise of the temperature occurs, producing a rapid evaporation of the material and the formation of the plume composed of ablated material. Commonly, in high pulsed energy ablation, the plasma plume produced at the target surface expands in the direction of the maximum pressure incline [9,10].

PED is a well-established technology to fabricate thin films for photovoltaic, superconductor, and optoelectronic applications [11,12]. PED technology belongs to the family of the channel spark discharges, in which a target material is ablated by the local heating induced by an accelerated electron beam. At the very beginning, this technique was mainly employed for the deposition of both inorganic, i.e., superconductive MBa₂Cu₃O₇-x, and organic, i.e., polytetrafluoroethylene (PTFE), thin films. The deposition applications of the technique expanded in the past years to include for example, biomedical materials, CuInGaSe₂ Solar Cells, In₂O₃ nano-films, LaMnO₃ thin Films, Poly (ethylene-co-vinyl acetate) films and nanostructured Ag thin films [13-17].

In this paper, the role of the gas type in the dynamics of the electron beam pulse system of the spark channel and the possibility of using the system for thin film deposition of carbon thin films is studied.

EXPERIMENTAL SETUP

Experimental setup for our channel spark Pulsed electron deposition system can be found in details in our previous publication [1]. The difference here is using of a commercial spark plug as triggering source for plasma generation. Figure 1 shows the experimental arrangement and the electrical circuit for coupling the negative high voltage signals to the device. The length of channel tube is 15 cm and the distance between hollow cathode and anode chamber is 5.5 cm. The 10 KΩ resistor was used to hold the discharge voltage at the main discharge tube and to ensure that the discharge is occurring first at this tube. Besides this resistor is (DC) blocking the capacitor discharge before the glow discharge is established at the main tube. O₂, N₂, and Ar gases in pressure range of 20–30 mTorr were employed as discharge gases. The applied voltage is varied between 9 and 13 kV with an external capacitance of 21.6 nF. Pearson coil and high voltage probe were used to monitor the discharge current and the voltage. Additionally, to investigate electron beam current characteristics a faraday cup was employed. An optical emission spectrometer (Ocean Optics USB4000 Spectrometer) was employed to monitor the reactive species during plume formation after bombardment of electron beam with the target (graphite).

EXPERIMENTAL RESULTS

1. Electric characterization

To study electrical characteristics of the system, the discharge current and voltage for O₂, Ar and N₂ as working gases were measured at different discharge voltage, pressure and capacitance. Figure 2 a, b, c shows a typical
discharge current and voltage waveforms for O\textsubscript{2}, Ar and N\textsubscript{2} respectively at pressure 20 mTorr and applied voltage -13 kV. When plasma reaches the cathode opening, capacitor starts discharging, so, the current quickly increases and the voltage drops down. In the hollow cathode discharge, the electrons have energy in same range of the accelerating voltage [18]. Note the N\textsubscript{2} discharge current is positive to reverse the Rogowski direction.

Figure 2 shows that, the current reached maximum value at 1110 A after a period of \(~\)466 nsec for Ar, \(~\)1189 A after a period of \(~\)343 nsec for O\textsubscript{2}, and \(~\)910 A after a period of \(~\)387 nsec for N\textsubscript{2}. The voltage started from (-13 kV) then the voltage falls to zero after 400 nsec.

Oscillations in the discharge current waveform were thought to be due to the
development of high-conductivity plasma channel between the ground electrode and the hollow cathode [19]. Current and voltage waveforms simulation were performed based on LCR circuit theory and shown in figure (3) for N₂ plasma. It should be noted that this simulation is only performed after the discharge of the capacitor on the beam acceleration section which can be simply modeled as a capacitor discharging in series with coil and resistor.

Based on this circuit theory current equation can be written as:

\[ I = I_0 e^{-\alpha t} \cos(\omega t) \]

Where, \( \omega = \sqrt{\frac{1}{LC} - \left(\frac{R}{L}\right)^2} \) is an angular frequency and \( \alpha = \frac{R}{2 \sqrt{LC}} \) is the damping factor.

![Fig. 3 Voltage and current waveforms simulations.](image)

It can be noticed from fig. 3 that, there is a good match between measured and simulated current waveforms. On the other hand, there is a mismatching between measured and simulated voltage waveforms due to the noise that picked up by the potential divider of bad isolation.

Variation of the maximum discharge current with applied voltage was calculated and shown in fig. 4 It was found that, there is a slight difference in maximum discharge current when using O₂, Ar, and N₂. So, the discharge characteristics may be independent of gas type [20].

![Fig.4: Dependence of maximum discharge current with the applied voltage for Ar, O₂ and N₂.](image)

Variations of the maximum discharge current and the total circuit inductance with operating gas pressure at varied discharge voltages for O₂ plasma were investigated and shown in fig. 5 (a, b) It is found that, with increase in both the pressure and discharge voltage the maximum discharge current increases. The maximum current was 1300 A at an applied voltage of -13 kV for Ar gas. On the other hand, because the device works in the left side of the Paschen curve, increasing the gas pressure leads to an increase in the maximum discharge current at a constant applied voltage [19]. By increasing the applied voltage more energy is pumped to the device, which results in an increase in the discharge current amplitude. The total inductance would be increased as both the pressure and the applied voltage decrease.

The rise in the pressure results in an increase in the plasma inductance, and consequently in a decrease in the periodic time of the current [10].
Two identical coils of 5 turns wounded at the same directions were installed at the acceleration tube to measure the magnetic properties of the accelerated electron beam. By using pick up coils electron beam speed was measured using time of flight method [19]. Figure 7 shows the measured electron beam speed was found to be about \(0.37985 \times 10^6\) m/sec and the corresponding value of beam kinetic energy was 4.1 eV. This value is greater than that obtained in our previous X-ray emission measurements which were less than 3 keV with efficiency of about 30% [1].

Plasma density due to beam acceleration can be calculated from maximum beam current and beam speed. It was found to be around \(1.96 \times 10^{30}\) m\(^{-3}\). As reported in [13], the current density \(J_{\text{eff}}\) of an electron beam is limited by the electron space-charge effect and is given by equation 1.

\[
J_{\text{eff}} = (2.3 \times 10^{-6})V_e^{3/2}/d^2 \quad \left( \frac{\text{A}}{\text{m}^2} \right) \quad (1)
\]

Where \(V_e\) is the acceleration voltage and \(d\) is the distance between the two electrodes.
3. Optical Characterization

Optical emissions spectra resulting from the interaction of the beam with the target (graphite) in presence of Ar and O\textsubscript{2} gases at applied voltage of 13 kV, measured from near the target surface are presented in Fig. 8 (a, b).

![Emission spectra for Ar and O\textsubscript{2} at 13 kV](image)

In figure 8 (a, b) we observe the formation of different ions like Ar\textsuperscript{+}, O\textsuperscript{+}, O\textsuperscript{++}, C\textsuperscript{+}, and C\textsuperscript{++} and different excited atoms and molecules like Ar, O\textsubscript{2}, C\textsubscript{2}, N\textsubscript{2}, and H (α, β, γ). At the presence of the Ar gas (see figure 8a) the electron beam continued to collision with the Ar atoms along the path between the end of the acceleration tube and target surface this produced the Ar atoms and ions, similarly with the O\textsubscript{2} gas (see figure 8b).

Carbon species were produced from the collision of the electron beam with the target surface. The ionic species that were produced from the collision deposit onto the substrate and produce a-C thin film. Also, we observe the formation of different species of N\textsubscript{2} (ions and excited molecules). These products were produced from the collision of the electron beam with N\textsubscript{2}. We attribute the presence of the N\textsubscript{2} to the base pressure. We also observe the formation of the H (α, β, γ). The source of the H\textsubscript{2} came from both the target material and the base pressure.

4. FTIR Measurements

The a-C:H films deposited over silicon substrate at different accelerating voltages between 9 to 13 kV for two different gasses O\textsubscript{2} and Ar show polymeric characteristics. FTIR spectra of the deposited a-C:H films at different experimental conditions are shown in figure 9. The spectra have narrow and well resolved vibrational bands and allow water to diffuse into the material along columnar assemblies as observed by typical H\textsubscript{2}O absorption bands around 1700 and 3250 cm\textsuperscript{-1}.

![FTIR spectra of the deposited a-C:H films at different experimental conditions](image)

As shown by fig. 9 one can find C-H deformation vibrations between 1300 to 1500 cm\textsuperscript{-1}, C=C stretching vibrations between 1500 and 1700 cm\textsuperscript{-1} from the carbon network, and C-H stretching vibrations around 3000 cm\textsuperscript{-1} [21-23].

The structure difference between the deposited films is due to difference in beam energy and current of Ar and O\textsubscript{2} as discussed before in the electrical characterization of the system. The films can be considered as C or a-C:H depending on the C-C sp\textsuperscript{3} bonds are saturated with some C-H sigma bond or the sp\textsuperscript{3} fraction is only related to pure C-C sigma bond. The last situation is mainly confirmed by the FTIR results as we will see later from fitting that some of C-C sp\textsuperscript{3} bond are saturated with hydrogen atoms. Obviously, the shape and the total intensity of the peak are quite sensitive to
the film structure. Soft, polymerlike hydrocarbon layers are characterized by a well-structured, intense IR absorption band. The CH vibrational bands can be considered as fingerprint for the type of a-C:H film. The changes of the microstructure of the deposited films. For all deposited films, it is clear that the density of \( \text{sp}^3 \)-CH\(_3\), \( \text{sp}^3 \)-CH\(_2\), \( \text{sp}^3 \)-CH groups, which contribute very strongly to the observed structure are very high while the density of \( \text{sp}^2 \)-CH\(_x\) groups are very low. The infrared absorption cross sections (often also called dipole strength) for the latter groups, for the \( \text{sp}^2 \)-related bands, is significantly lower than for the \( \text{sp}^3 \)-CH\(_3\) groups. In order to obtain both \( \text{sp}^2 \) and \( \text{sp}^3 \) fraction and hence know more information about the films structure 

\[
E_g = 4.59 x^2 - 0.561 x + 0.539
\]

In this equation \( x \) is the \( \text{sp}^3 \) content, and \( E_g \) is the optical band gap. The relation between \( E_g \) and \( H \) content in the samples can be obtained through the following relation.

\[
H = (E_g + 0.9)/9
\]

The \( E_g \), and \( H \) content estimated according to equation 1,2 are listed in Table 1 for all deposited films.

Comparison between the structure of the films deposited for \( \text{O}_2 \) and \( \text{Ar} \) as feeding gasses at two different charging voltage of 9 and 13 kV is shown in Table 1.

Fig. 10: Area estimation of \( \text{sp}^4 \) CH\(_x\) and \( \text{sp}^2 \) CH\(_x\) contributions of the deposited a-C:H films at different experimental conditions.
CONCLUSION

In this paper, the gas type role on the dynamics of channel spark pulsed electron deposition system was studied. Electrical, magnetic, and emission properties of channel spark system were examined for oxygen, argon, and nitrogen gases. Discharge current and voltage waveforms are best described and simulated using LCR circuit theory. The discharge characteristics may be independent of gas type. With increase in both the pressure and the discharge voltage the maximum discharge current increases. The presence of carbon excited species in the optical emission spectra confirmed the interaction of the electron beam with the graphite target. The possibility of using the electron beam in the deposition of carbon thin films is demonstrated by the deposition of a-C:H thin films on silicon substrates as confirmed by FTIR measurements.

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Table 1 comparison between the structures of the deposited films at different conditions.

| Samples | SP3 % | SP2 % | Eg (eV) | H % |
|---------|-------|-------|---------|-----|
| 9kV Ar  | 63.61775 | 36.38225 | 2.04041 | 32.67126 |
| 9kV O₂  | 56.15942 | 43.84058 | 1.67214 | 28.57932 |
| 13kV Ar | 65.50758 | 34.49242 | 2.14184 | 33.7982 |
| 13kV O₂ | 63.31512 | 36.68488 | 2.02448 | 32.4918 |


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