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Influence of O_2 and N_2 Concentrations on the Characteristics of Plasma in DC Cylindrical Magnetron Discharge by Langmuir Probe

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Abstract

Using the Langmuir probe method, the reactive plasma parameters were studied in different ratios of oxygen and nitrogen concentrations in a DC cylindrical discharge device.The plasma parameters such as plasma potential, electron density and electron temperature wereextracted from the current-voltage characteristic's curve of Langmuir probe to find the optimum conditions for deposit the oxynitride thin films.Chromium thin films were exposed to various O2/N2 partial pressures to obtain optimum valuefor produce of oxynitride chrome thin films. In addition, the influence of the magnetic field on the structural properties of the chrome oxynitride thin films was obtained. It is observed that for equal percentage of reactive gases, the optimum condition of the plasma discharge takes place in which the crystalline phases of oxynitride chrome thin films appear.

Keywords: *plasma discharge, Langmuir probe, magnetron sputtering, oxynitride thin films, plasma parameters.*

Introduction

Developments of plasma's applications in industry causes to many researchers have beenfocus on it. Plasma etching, plasma deposition, implantation andsurface modification for producing films on surfaces are some examples of plasma applications in industry [1-2]. Deposition by means of plasma sputtering is acommon technique for coating and is used to create new materials. It is known that, the basic properties of thin film produced by plasma such as film composition, crystal phase and orientation, film thickness and microstructure are influenced by ion's

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behavior and deposition conditions [3-4].

However, the mixture of reactive working gasses in the discharge change some of the characteristic features of deposited layer. Oxygen and Nitrogen discharges are widely used for industrial materials, such as, production of photoresist material, deposition of oxide or nitride films and functionalization of polymers [5-6]. Also Nitrogen is used for deposition of nitride layers to prevent corrosion of metals. Because of wide utilization range of Nitride or Oxide layer deposited by means of the plasma magnetron in industry, there are many experimental researches that focus on plasma diagnostics in electronegative discharges of N_2 or O_2 as working gases [7-8]. There are some methods to deposit oxynitride thin films in which the magnetron sputtering as a controlled deposition technique has been used to deposit uniform and homogeneous thin films.

On the other hand, the oxide, nitride and also oxynitride of transition metals are of great interest because of their electrical, opticalproperties and their inert chemical behavior. Especially, oxynitrides have crystalline structure formed by a mixture of ionic and covalent bonds between oxygen/ nitrogen and metal that provides them with physicochemical properties different from those exhibited by oxides and nitrides [9]. There have been many preparation methods for aoxynitride thin film such as reactive planar sputtering in oxygen/nitrogen gases, oxidative annealing, or metal organic chemical vapor deposition [10-12]. However, among the plasma methods to produce the oxynitride thin films, there are no investigations on the formation of such films by means of *cylindrical* plasma sputtering. In planar magnetron, permanent magnets create the magnetic field and it is impossible to change the magnetic field intensity while in cylindrical magnetron device, a coil generates magnetic field and experiment can be performed in different magnetic fields intensities. In addition, the geometry of the magnetron has large influence on plasma characteristics. Therefore, the results of the present paper vary considerably from planar magnetron.

Regarding to importance of the magnetic field intensity, it is to be noted that by using an external magnetic field, one can better control different features of the discharge such as energy of the impactthe ions and ion flux. A better understanding of the discharge behavior under the influence of an applied magnetic field is therefore quite necessary.The magnetic field in these systems can be either non-homogeneous created typically by permanent magnets like planar unbalanced magnetron or almost homogeneous created by coils like a cylindrical magnetron.

In this paper, a cylindrical magnetron dischargein the presence of O_2/N_2 is investigated by Langmuir probe to obtain the optimum value of relative concentration of O_2/N_2 and also the optimum value of the applied magnetic field.The cylindrical magnetron discharge has two distinguished feature: a nearly uniform magnetic field and a variable magnetic field. Different ratios of Oxygen and Nitrogen are used in constant pressure and discharge power in different of magnetic fields intensity. The plasma parameters such as plasma potential, electron density and electron temperature are obtained via the current-voltage characteristic's curve of Langmuir probe. As we are going to obtain the optimum value of the magnetic field and relative concentration of the oxygen to nitrogen toobtain oxynitride thin films, the chrome thin films has been chosen to exposed to the reactive plasma. Finally, the influences of O_2/N_2 partial pressure and also the magnetic field on the structure of the chrome oxynitride thin films are investigated. The structure of the paper is as follows. Following the introduction we present the experimental details for the preparation and then the results are discussed in Sec.4. We conclude the paper in final section.

Experimental

We investigated the influence of O_2/N_2 relative concentration and magnetic field on plasma characteristic via the Langmuir probe in constant pressure. The experimental setup consisted of two coaxial cylindrical electrodes as cathode (inner one) and anode (outer one) with 3 and 12 (cm) in diameter and 13cm length respectively. The system was evacuated down to torr by means of a mechanical and diffusion vacuum pumps. A nearly uniform magnetic field, parallel to the axis of the cylinders, was generated by a coil (120 turns) around the outer cylinder.The working of gases with different ratios of O_2 and N_2 concentrations were introduced in to the chamber at torr. It is to be noted that the experiments are performed at constant voltage mode equal to 640 (V). The Langmuir probe consisted of quartz tube with cylindrical tungsten tip with 5mm length and 0.5mm diameter was posited at the middle of electrodes. Probe tips made of tungsten were easily etched. Probe was connected to a DC power supply and positively and negatively biased, so current of plasma was measured at various applied voltages. For different ratios of N_2 and O_2 gasses and from the I-V characteristics of probe, the plasma parameters were obtained in different magnetic field [13]. In the first step, the plasma parameters have been obtained via the Langmuir probe to obtain the optimum value of magnetic field and ratios of N_2 and O_2 gasses. In the second step the chrome thin film were obtained by magnetron sputtering. The argon (Ar) (99.995% pure) gas was used as sputtering gas and the chrome (99.99% pure) with 10 cm diameter was employed as a target. The distance between the target and substrate was about 6cm. The deposition chamber was evacuated down to a base pressure of 2×10-5 mbar using combination of diffusion and rotary pumps and after introducing the Ar gas into the deposition chamber, the pressure was maintained at 8×10^{-2} mbar. For all chrome thin films, the deposition time was kept at 3 min. In last step, the as-deposited chrome films were exposed to the plasma in the cylindrical magnetron in the presence of different ratios of oxygen-nitrogen gas mixtures .At the last step, the exposed films were characterized by the X-ray diffraction (XRD) (Philips, PW 3710, Cu Kα radiation λ=0.154056 nm) for analyze the crystallographic structure and orientation of the films. The surface morphology and microstructure of films w analyzed by atomic force microscopy (AFM) (Auto probe cp, Park scientific instrument).

Result and discussion

In Figure 1, the discharge current as function of oxygen and nitrogen concentrations is shown for three values of magnetic fields intensity at constant pressure of torr in constant voltage mode. It shows that by amplifying the magnetic field intensity, the current discharge increases while there is a minimum value in discharge electric current for the middle range of relative oxygen concentration. Since the resistivity of the plasma depends on the applied magnetic field,by rising up the magnetic field intensity, the probability of ionizing collisions increases and in the fixed voltage mode, the discharge current increases.

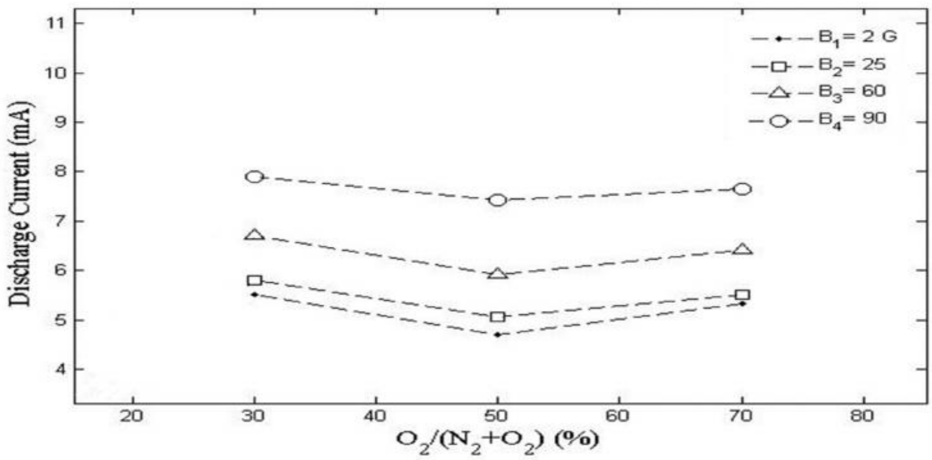

Figure 1. Discharge current vsoxygen/nitrogen concentrations for different values of magnetic field intensity.

In Figure 2, the variations of plasma potential as function of O_2/N_2 concentration is shown in different magnetic fields intensity. According to this figure, the plasma potential decreases by increasing the nitrogen concentration. As we have shown numerically in Ref. [14], in electronegative discharge, there is a inverse relationship between sheath thickness and negative ions concentrations and also the plasma potential increases with broadening the sheath thickness. By increasing the relative oxygen concentration, sheath thickness or plasma potential decreases. In addition, applying a magnetic field leads to decrease of sheath thickness and plasma potential[15].

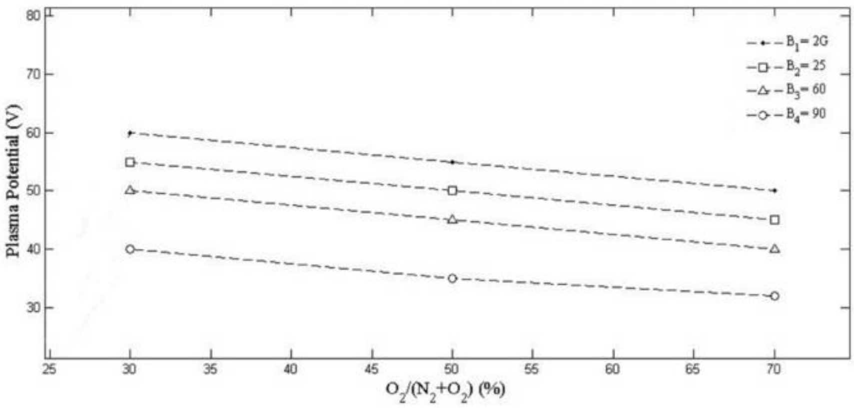

Figure 2. Plasma potential vs oxygen and nitrogen concentrations for different values of magnetic field intensity at fix pressure.

In Figure 3, electron temperature is plotted as a function of concentration of working gasses in different of magnetic fields. By rising up the O_2 concentration, the electron temperature decreases at constant voltage mode. However, in the presence of high concentration of oxygen, the influence of the magnetic field disappears which is a consequence of decrease of the electron temperature and high electronegativity of the oxygen.

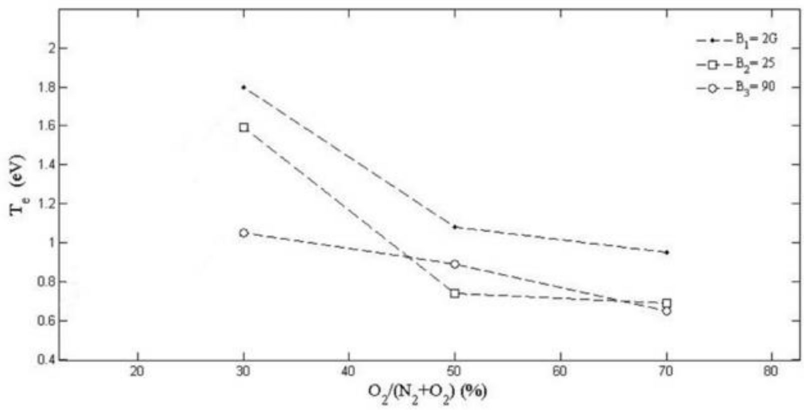

Figure 3. Electron Temperature vs oxygen and nitrogen concentrations for different values of magnetic field intensity at fix pressure.

Figure 4 shows electron density as function of magnetic field intensity for three values of working gas percentages in fix pressure. As it can be seen, there is an optimum value for magnetic field that electron density is in its maximum value. In high magnetic field, the electronis confined in vicinity of the cathode followed by decrease of electron density according to Ref. [16]. On the other hand, electron density has maximum value for same percentage of N_2 and O_2.

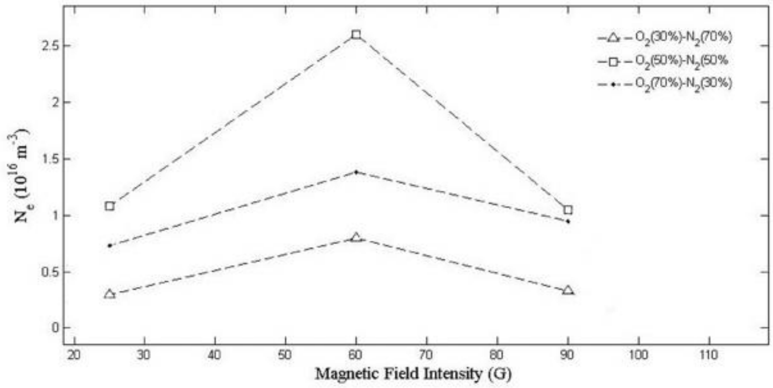

Figure 4. Electron densityvsmagnetic field intensity fordifferent values of oxygen and nitrogen concentrations at fix pressure.

Figure 5 shows the variation of electron density and electron temperature as function of concentration of working gases at constant pressure in the absence of magnetic field. By increasing the oxygen concentration, electron density and electron temperature decreases. The decrease of the electron density is a result of the higher electronegativity of the oxygen respect to the nitrogen. This result is in agreement with the results of the discharge containing the oxygen and argon [16].

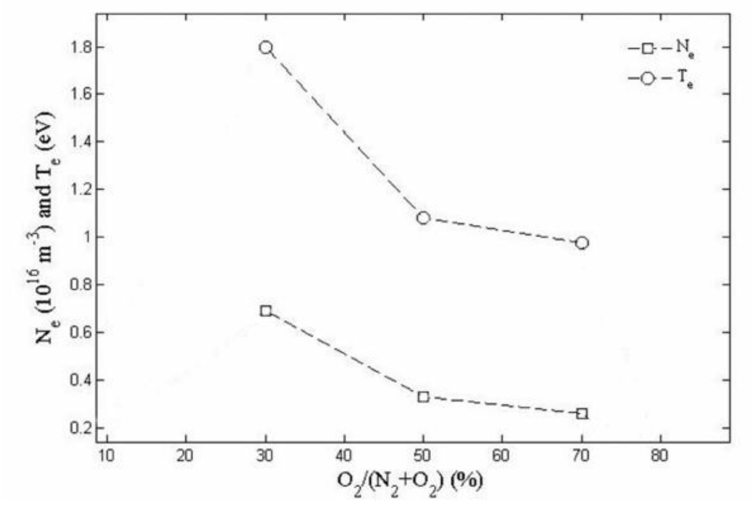

Figure 5. Electron Temperature and electron densityvs oxygen and nitrogen concentrations in the absent of magnetic field.

Next, the influence of the plasma parameters on the interaction of plasma-thin films was investigated. To obtain the influence of the plasma discharge on the thin film characteristics, the chrome thin films are located on the cathode of the discharge and are exposed to the plasma treatments with different ratios of O_2/N_2 concentrations. In the figure 6 the XRD patterns of the chrome thin films exposed to the plasma discharge are shown for three values of O_2/N_2 concentrations.

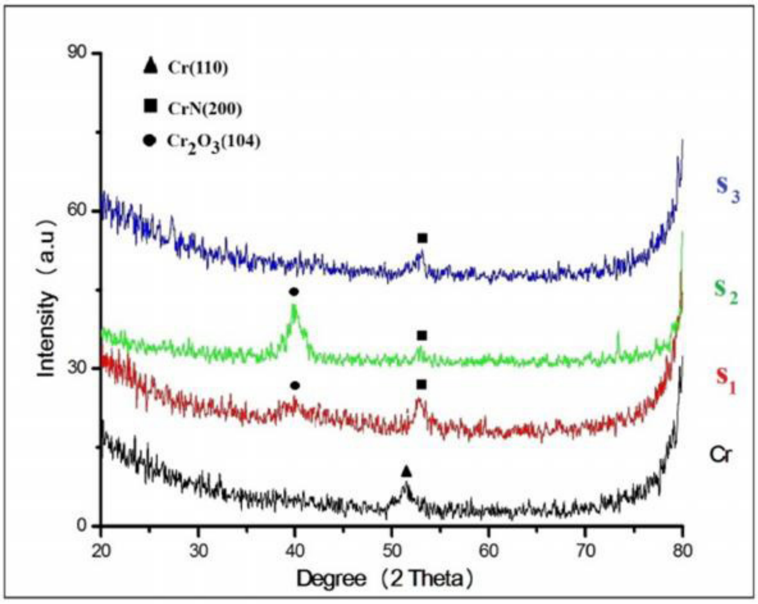

Figure 6. X-ray diffraction patterns of (Cr) and S_1) 30% oxygen – 70% nitrogen ,S_2)50% oxygen – 50% nitrogen and S_3) 70% oxygen – 30% nitrogen.

It can be seen that in the presence of the equal gas mixture of oxygen and nitrogen, the interaction of the discharge with the chrome layers leads to formation of Cr2O3and CrN simultaneously. In addition, from the figure 7, it is observed that the rms roughness of the layers is in its maximum values in the presence of the 50% oxygen – 50% nitrogen.

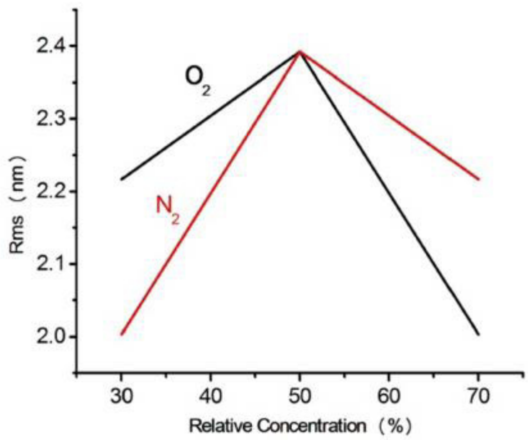

Figure 7. The roughness of the chrome layers exposed to the different ratios of the O_2/N_2 concentrations.

Conclusion

Influences of nitrogen and oxygen concentration as reactive working gases on plasma parameters have been investigated for different of magnetic fields intensity in a DC cylindrical discharge by the Langmuir probe technique. The results show that discharge current decreases in the equal ratio of O_2/N_2 concentration. In addition, the plasma potential and electron temperature decreases by increasing of oxygen concentration. The magnetic field gives rise to reduction in plasma potential and electron temperature. It was observed that, the maximum value of electron density takes place for same ratio of N_2 and O_2. In addition, for the same percentage of the reactive gases, the treatment of chrome layer by reactive plasma leads to formation of oxynitride thin film. In this work, all the chrome thin films have the same thickness. Therefore, for future work, it is needed to consider more thin films with different thicknesses to investigate the influence of the plasma parameters on the chemical composition of thin films surfaces.

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