Effect of (O2/Ar) Different Mixing Ratios in a Glow Discharge Plasma on the Structural Properties of Niobium Oxide Film

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Abstract

Niobium Oxide (Nb_2O_5) films were prepared on glass substrates using a plasma sputtering system with different ratios of (O_2/Ar) gas mixture. The annealing process of all samples was done at (650 °C) for one hour. The X-ray diffraction (XRD) results of the Niobium oxide (Nb_2O_5) films of ratios in group-1, group-2 and group-3 show that the phase of Niobium Oxide was not present before and after annealing, due to the decrease in the ratio of oxygen gas compared to the ratio of Argon gas. While the X-ray diffraction (XRD) of Niobium oxide films of ratios in group-4 and group-5 indicated the phase of Niobium Oxide (Nb_2O_5) after the annealing because the heat treatment led to the crystalline uniformity of the layer of Niobium oxide and thus the growth of the phases. Also, the X-ray diffraction (XRD) of (Nb_2O_5) films of ratio in group-6 demonstrates the presence of multiple and clear phases of Niobium oxide (Nb_2O_5) before and after annealing, as a result of the large increase in the proportion of oxygen gas compared to the proportion of the Argon gas, which led to the growth of the phases and the largest crystalline system.

Key Words: Niobium Oxide, Plasma Sputtering, and (Oxygen / Argon) (O_2/Ar) Mixture

تأثير نسب الخلط المختلفة لغازي (O2/Ar) في بلازما التفريغ المتوهج على الخصائص التركيبية لغشاء أوكسيد النيوبيوم

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الخلاصـــة

حُضرت أغشية أوكسيد النيوبيوم (Nb_2O_5) على قواعد من الزجاج بأستخدام منظومة بلازما الترذيذ وبنسب مختلفة من خليط لغازي الأوكسجين/الأركون (O_2/Ar)، وتمت عملية التلدين للنماذج بدرجة حرارة (650 °C) لمدة ساعة واحدة، حيث بينت فحوصات حيود الأشعة السينية (XRD)للأغشية في المجموعة-1 والمجموعة-2 والمجموعة-3 عدم ظهور طور أوكسيد النيوبيوم قبل وبعد التلدين بسبب انخفاض نسبة غاز الأوكسجين مقارنة بنسبة غاز الأركون، بينما أظهرت فحوصات حيود الأشعة السينية للأغشية في المجموعة-4 والمجموعة-5 ظهور طور (Nb_2O_5) بعد التلدين وعدم ظهوره قبل التلدين لأن المعاملة الحرارية أدت الى الانتظام البلوري لطبقة أوكسيد النيوبيوم وبالتالي نمو الأطوار. كما أظهرت فحوصات حيود الأشعة السينية للأغشية في المجموعة-6 وجود أطوار متعددة وواضحة لأوكسيد النيوبيوم (Nb_2O_5v) قبل وبعد التلدين نتيجة الزيادة الكبيرة لنسبة غاز الأوكسجين مقارنة بنسبة غاز الأركون والذي أدى الى نمو الأطوار وأنتظام بلوري أكبر.

الكلمات المفتاحية: أوكسيد النيوبيوم، بلازما الترذيذ وخليط الأوكسجين/الأركون (O_2/Ar)

Introduction

Plasma sputtering method is one of the most well-known procedures that have been broadly utilized for deposition of thin films in material processing. It offers two fundamental preferences. Firstly, the simplicity with which one can deposit on wide region because of the great plasma consistency. Secondly, the deposition amount is high, (Bouchikhi, *et al*., 2012). The utilization of techniques about plasma has been effectively used since the 60s in various fields in material science, (Li, *et al*., 1997). The impact of nature and substrate structure on physical and chemical properties and surface morphology are noteworthy, (Gholamali, *et al*., 2018). Gas is typically an electric insulator. But, when an adequately enormous voltage is connected over a gap containing a gas or gas mixture, it will breakdown and conduct electricity, the reason is that the electrically neutral atoms or molecules of the gas have been ionized into negatively charged electrons and positively charged ions. The idea of the breakdown and the voltage at which this happens changes with the gas species, gas pressure, gas flow rate, the separation distance of the electrodes, the nature of the high voltage supply (for example DC, AC, radiofrequency or microwave) and electrical hardware. The resulting ionized gas is often called a discharge or plasma. (Shahidi, *et al*., 2012). There are many studies to utilize plasmas, such as (Lee, *et al*, 2012). study that Helium (He) gas was utilized to produce plasma at a fixed flow rate of 4 liters per minute, (O_2) was added to the (He) to watch the impact of the gas mixture. The outcomes demonstrate that the dielectric barrier discharge DBD plasma system has the potential for use in sanitizing food products. (Mishra, *et al*., 2016), used a cold plasma to study one of the newest technologies used in food industry for microbial inactivation, it is crucial that foods are supplied without any microbial contamination as many products are eaten raw, without loss of physicochemical or sensory properties. (Sproul, *et al*., 2005), show that the method of reactive gas control during reactive sputtering strongly influences the deposition rate and film properties of the compound being deposited. (Chen, *et al*., 2017). show that the results of the simulation and the experiment are performed for different power and oxygen mole ratios and a good qualitative and numerical agreement is obtained. (Ogawa, *et al*., 2014). found that the functionalization with the plasma discharge created with the mixture gas, Argon (Ar)/Nitrogen (N_2)/Hydrogen (H_2), was the most effective in dispersing of multi-walled carbon nanotube (mwcnts) in tetrahydrofuran (THF) than the plasma created with the other two mixture gases, Argon/Oxygen or Argon/Nitrogen. The objective of this research is to study the structural properties of (Nb_2O_5) and discharge behaviour in different (O_2//Ar) gas mixtures on the Niobium oxide films.

Materials and Methods

Experimental

The Niobium oxide films (Nb_2O_5) were prepared on a substrate of glass with dimensions of (2 cm x 2 cm) using the plasma sputtering system as described in Figure (1).

The plasma sputtering system consists of a stainless-steel vacuum chamber with a length of 42 cm and a diameter of 40 cm. The discharge operates in a DC power supply (Maximum Voltage 5 kV, 30 mA), and digital multi-meters were used to measure the discharge current and the voltage.

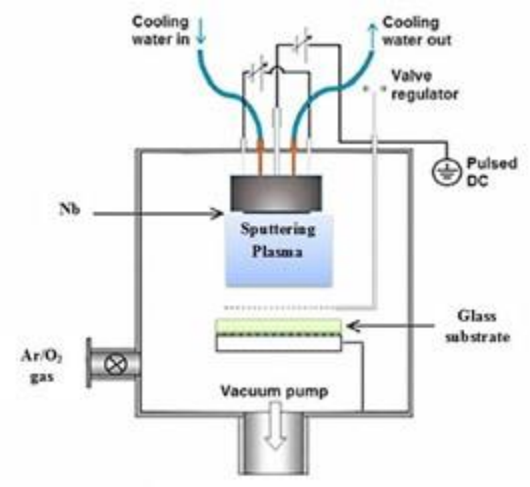

Fig. (1) Schematic of Plasma Sputtering of (Nb_2O_5) Films.

Inside the chamber there are two electrodes (Anode and Cathode), the diameter of each of them is 12 cm, and the separation was fixed at 4 cm between them, the glow-discharge is formed. Also, a pressure gauge (Type VD84-tHyr-cont) was used to measure the gas pressure inside the vacuum chamber and a thermometer (MTD-96, Maxwell) was used for measuring the temperature. The plasma sputtering system has two stages rotary pump (module DS-602, Italy) and turbopump (Pfeiffer-balzers). Niobium metal was used as cathode with purity (99.95%), and the mixture of O_2/ gas and Ar gas with purity (99.99%). Niobium oxide films were deposited on glass substrates with different rates of (O_2/Ar) mixture as shown in Table (1). The pressure inside the chamber is 3x10-3 mbar, and the temperature increasing with increasing the time of operation in range (100-120 °C), the applied voltage was 3 kv and the current was 10 mA for all samples. The Figure (2-A) shows the plasma sputtering of (V) film of ratio (O_2: Ar = 1.5×10^{-2}: 0.5×10^{-2}) (group-4), when O_2 concentration more than Ar concentration, therefore the color of glow tends to the violet and lavender. While the Figure (2-B) shows the plasma sputtering of (Nb_2O_5) film ratio (O_2: Ar = 0.5×10^{-2}: 1.5×10^{-2}) (group-2), when O_2 concentration less than Ar concentration, therefore, the color of glow tends to the violet pale lavender blue.

Table (1) The Rate of (O_2//Ar) Mixture.

Name of group	O_2 (10^{-2}) L	Ar (10^{-2}) L
group-1	1	1
group-2	1	1.5
group-3	1	2
group-4	1.5	0.5
group-5	2	1
group-6	2.5	0.5

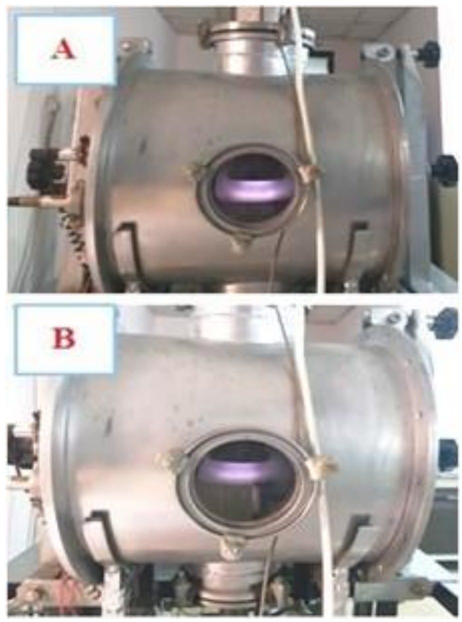

Fig. (2) (A) Plasma Sputtering of (Nb_2O_5) Film for Sample from Group-4. (B) Plasma Sputtering of (Nb_2O_5) Film for Sample from Group-2

The samples of Niobium oxide films were annealed at (650 °C) for one hour, and the structure of the Niobium oxide films before and after heat treatment was evaluated by X-ray diffraction (XRD) in (2θ) configuration using (Cu Kα) radiation in a Philips X'PERT instrument.

Table (2) The Values of (2θ) and (hkl) of (Nb_2O_5)

2θ degree	Intensity (cps)	(h k l)	2θ degree	Intensity (cps)	(h k l)
10.524	3	2 0 2	44.902	6	2 11 0
14.037	5	2 0 3	44.996	6	3 3 0
15.827	2	3 0 3	45.715	1	1 14 0
16.971	5	1 3 0	46.059	25	0 0 2
18.126	1	0 6 0	48.789	6	1 13 1
22.606	90	0 0 1	49.727	16	0 16 0
28.309	100	1 8 0	50.433	1	3 2 1
28.870	45	2 0 0	50.885	25	3 8 0
29.062	15	2 1 0	54.971	25	1 8 2
32.778	5	2 5 0	55.294	20	1 17 0
34.357	3	2 6 0	55.330	20	2 0 2
34.466	1	1 7 1	55.622	3	2 15 2
36.495	50	1 8 1	56.327	15	3 8 1
36.993	20	2 0 1	58.559	10	2 16 0
37.120	10	2 1 1	59.938	4	4 0 0
42.506	3	2 10 0	60.024	4	4 1 0
42.653	4	1 13 0	60.897	3	3 13 0

Results and Discussion

The values of (2θ) and the miller indices (hkl) of (Nb_2O_5) according to the ASTM pdf-number (431042) are shown in Table (2). The X-ray diffraction (XRD) examinations of the Niobium oxide (Nb_2O_5) films, that deposition by utilizing plasma sputtering system at different ratios of (O_2/Ar) gas mixture, for groups (1, 2, and 3) in Table (1) show that not the appearance of the phase of Niobium oxide (Nb_2O_5) before and after annealing at (650 °C) and at the same conditions of operation of plasma sputtering are amorphous phases as shown in Figures (3, 4 and 5).

This is due to the decrease in the ratio of oxygen gas compared to the ratio of Argon gas and to the fact that the sputtering layer is very low. The X-ray diffraction (XRD) of Niobium oxide (Nb_2O_5) films for groups (4 and 5) in Table (1) indicated the appearance of the polycrystalline phases of Niobium Oxide (Nb_2O_5) after the annealing at (650 °C), but not appear before the annealing as shown in Figures (6 and 7).

Due to the annealing operation which led to the growth of the phases as a result of regularity crystalline layer of Niobium oxide (Nb_2O_5) and increasing the proportion of oxygen gas.

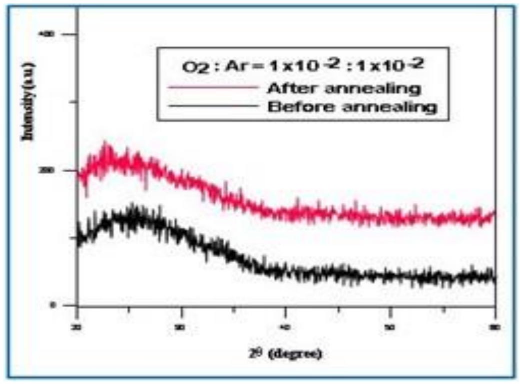

Fig. (3) The X-Ray Diffraction (XRD) Patterns Obtained for (Nb_2O_5) Films in Plasma Sputtering at (O_2/: Ar =1x10-2: 1x10-2), group-1.

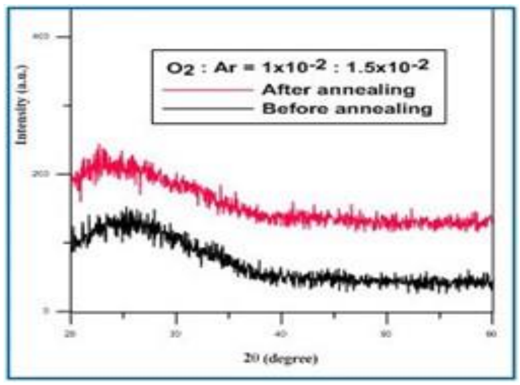

Fig. (4) The X-Ray Diffraction (XRD) Patterns Obtained for (Nb_2O_5) Films in Plasma Sputtering at (O2: Ar =1x10-2: 1.5x10-2), group-2.

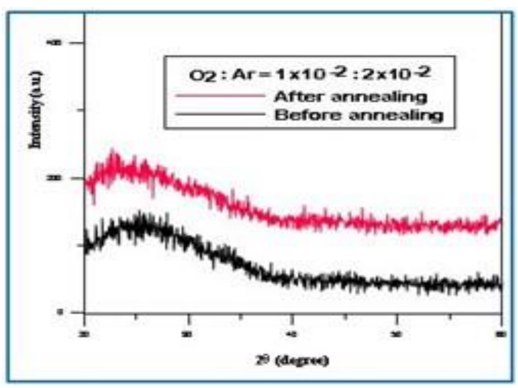

Fig. (5) The X-Ray Diffraction (XRD) Patterns Obtained for (Nb2O5) Films in Plasma Sputtering at (O2/: Ar =1x10-2: 2x10-2), group-3

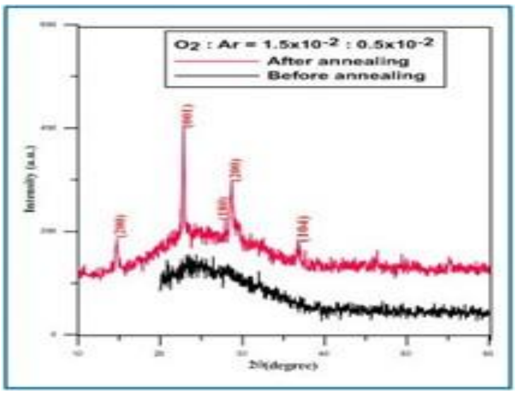

Fig. (6) The X-Ray Diffraction (XRD) Patterns Obtained for (Nb_2O_5) Films in Plasma Sputtering at ($O2:Ar=1.5x10^{-2}: 0.5x10^{-2}$), Group-4.

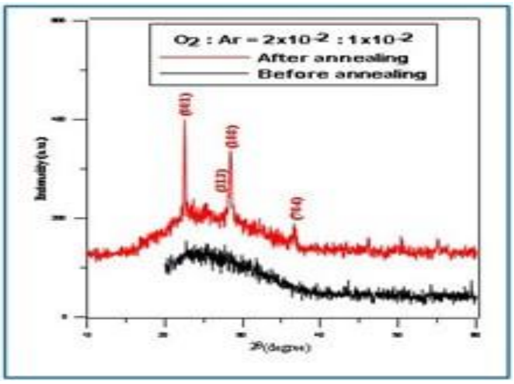

Fig (7) The X-Ray Diffraction (XRD) Patterns Obtained for (Nb2O5) Films in Plasma Sputtering at (O2:Ar =2x10-2: 1x10-2), Group-5

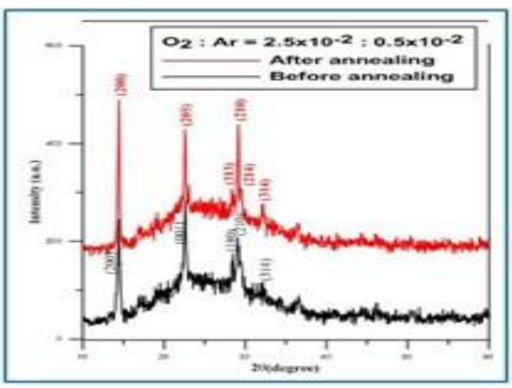

Fig (8) The X-Ray Diffraction (XRD) Patterns Obtained for (Nb_2O_5) Films in Plasma Sputtering at (O2/: Ar =2.5x10-2: 0.5x10-2), Group-6

Also, the X-ray diffraction (XRD) of Niobium oxide (Nb_2O_5) films for group-6 in Table (1) demonstrates the presence of multiple and clear phases (Polycrystalline) of Niobium oxide (Nb_2O_5) before and after annealing at (650 °C), as in Figure (8).

The annealing process led to greater crystalline growth and uniformity of Niobium oxide (Nb_2O_5) films. We can conclude that increased oxygen gas compared to the ratio of Argon gas and annealing operations leads to good thin films and good crystalline growth, so it can be used in many applications, such as in solar cells (Chen, *et al*., 2001), and in biomedical applications (Amaravathy, *et al*., 2014).

Conclusions

Plasma sputtering is one of the most important techniques for the preparation of films. The experiment has shown that a ratio of the (O_2/Ar) gas mixture, which has an oxygen gas ratio greater than the Argon gas ratio ($O_2:Ar = 2.5\times10^{-2}:0.5\times10^{-2}$) for the preparation of the Niobium oxide film (Nb_2O_5), leads to the best film due to appearance of the phases clearly and as demonstrated by X-ray diffraction examinations of the film. In addition, the annealing of the prepared film gives good results because the heat treatment led to regularity of the crystalline structure of the film, which is due to the appearance of the phases.

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