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Characterization of the Copper Oxide Thin Films Deposited by DC Sputtering Technique

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ABSTRACT

Nanocrystalline Copper Oxide films were deposited on glass substrates by plasma dc sputtering. The effected of discharge current on the structural and optical properties of sputtered films were studied .X-ray diffraction peak of Cu_2O (111) and Cu_4O_3 (112) direction was observed at discharge current of (15-30) mA when annealed at 500 0C for 2 h. The optical energy gap for the prepared films is estimated to be in (2.05- 2.3) eV range. It was found that the effect of preparation conditions on thin films thickness strongly depends on the discharge current of argon plasma.

دراسة خصائص اغشية اوكسيد النحاس المحضرة بطريقة الترذيذ للتيار المستمر

الخلاصة

تم في هذا البحث تحضير أغشية نانويه من مركبات اوكسيد النحاس رسبت على قواعد زجاجية باستخدام الترذيذ بالبلازما . تم مناقشة تاثير تيار التفريغ على الخصائص التركيبية والبصرية وباستخدام حيود الأشعة السينية والتي أوضحت وجود التركيب متعدد التبلور. ظهرت في الأغشية المحضرة بتيار تفريغ يتراوح بمدى mA (15-30) والملدنة بدرجة حرارة $500\ ^0C$ لمدة ساعتين وجود مستويات (111),(112) للاطوار(Cu_2O,Cu_4O_3). كما وجد إن فجوة الطاقة البصرية للأغشية المحضرة تتراوح من eV (2.05-2.3). كما وجد إن سمك الأغشية المحضرة يتأثر مع تيار التفريغ لبلازما الاركون.

INTRODUCTION

Copper Oxide Cu_2O; is a material of interest in the optoelectronic devices band on oxide semiconductors. Cu_2O has a direct band gap energy 2.1 ev [1,2]; is abundantly available, nontoxic in nature with low cost of production. The p-type conductivity arises due to copper vacancy (V_{Cu}); which is reported to form acceptor level at 0.12- 0.70 eV [3] above valance band. Cu_2O thin films find many applications in hetrojunction, solar cells, electro chromic devices, and Oxygen and humidity sensors [4,5]. Copper oxide is a semiconductor which shows varying optical behavior because of stoichiometric deviations arising from its methods of preparation and parameters [6]. Thin films of Cu_2O have been fabricated using the sputtering method[7,8], different techniques such as molecular beam epitaxial[9], chemical vapor deposition[10] ,reactive rf, and dc magnetron sputtering[11]. Among all the

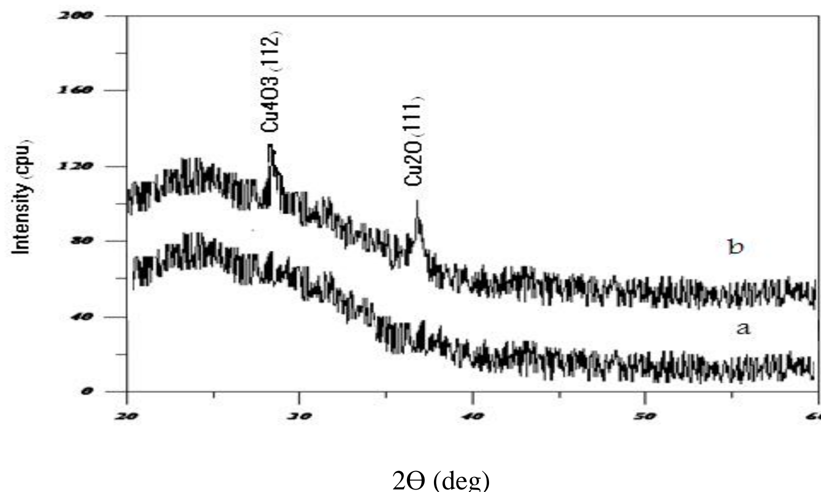
techniques of deposition, direct current (DC) magnetron sputtering is one of the best compositions. The physical properties of the Cu_2O films are influenced not only by deposition techniques, but also by process parameters, such as argon partial pressure, sputtering power and sputtering pressure. In this study the Cu_2O thin films were prepared on glass substrate by DC magnetron sputtering and the influenced of argon partial pressure and related the discharge current and on the structural and optical properties of the films were investigated.

EXPERIMENTAL PROCEDURES

The films were dc sputtered from copper target (99.9) in a homemade plasma system. The sputtering chamber was evacuated to less than 5×10^{-5} mbar, throttled and the sputtering gas pressure was 2×10^{-1} mbar with a discharge current (15-30) mA ,and voltage (0.6-1) KV .The glass substrates with dimensions (4×1.5×0.15) cm, and optical transmission of about 95% were used for depositing the films. The optical spectrum of absorption and transmission were measured by the (UV-VIS-2601, Biotech Engineering Management CO.LTD) spectrophotometer. X-ray diffraction (XRD) was used to characterized the structure of the films on a diffractometer of (Shimadzu) with Cu Kα (λ=0.15418 nm) radiation. The atomic force microscopy (AFM) is contact mode was used to analyze the morphological feature on Ingstrom Inc, (AA3000).

RESULT AND DISCUSSION

Figure (1) presents the XRD diffraction patterns in the range of 2θ where the diffraction patterns for the deposited samples; where Figure (1-a) shows the amorphous structure, and Figure (1-b) shows the presence of diffraction peaks (111) ,and (112) of (Cu_2O) and (Cu_4O_3) phase respectively ;for the deposited samples which annealed at (500 ^{0}C) . Bhattacharyya *et al.* [12] also obtained Cu_2O samples with reflection from (111) ,and (112) planes of cubic phase by RF sputtering technique ,and table(1) lists the observed of Cu_2O of plane (111) besides the trace amount of Cu_4O_3 of plane (112) which appeared because of excessive oxygen. This effect result due to the annealing process.



2Θ (deg)

Figure (1) XRD Patterns of sputtered copper oxide films (a) as deposited, and (b) annealed at 500 ^{0}C.

Table (1) X-ray diffraction data for copper oxide films.

2-θ (degree)	hkl	d (A^0)
37.009	111 (Cu_2O)	2.427
28.090	112 (Cu_4O_3)	3.174

Figure (2) shows the AFM images of four specimens surface for different films thickness; due to changing the current discharge at argon plasma. The most important feature that we can observe from the images is decorations of collapsed grains are distributed homogeneously with a pattern consistent with a polycrystalline film. A quantitative analysis of the roughness from the images is summarized in Table (2) here the normalized roughness ρ of the films is presented as a function of the film thickness.

Table (2) Surface roughness of copper oxide thin films.

sample	Thickness (nm)	Discharge current I_d (mA)	Normalized roughness (ρ)
A	130	15	12.3
B	140	20	26.3
C	145	25	15.7
D	150	30	27.8

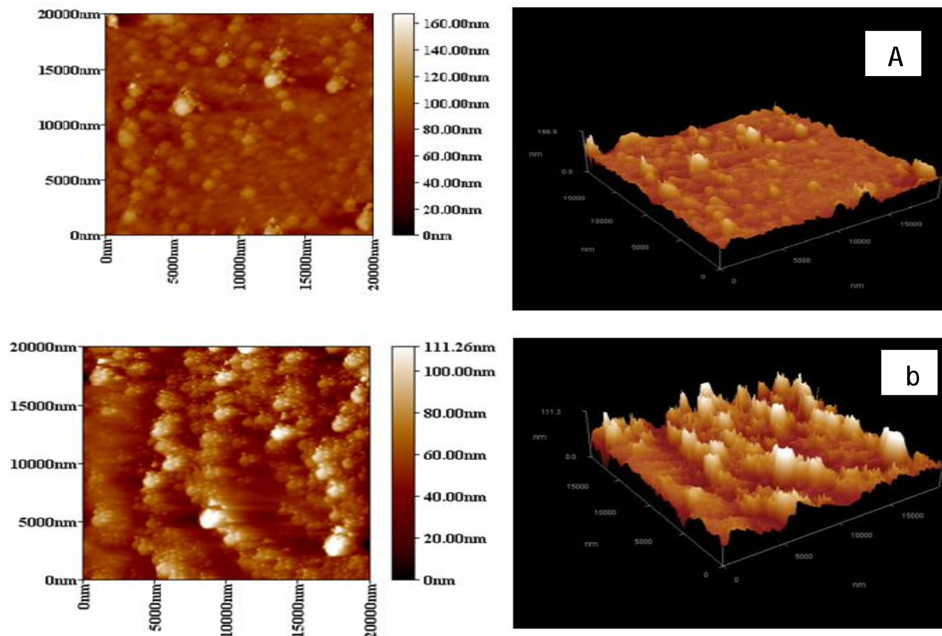


Figure (2) to be Continued

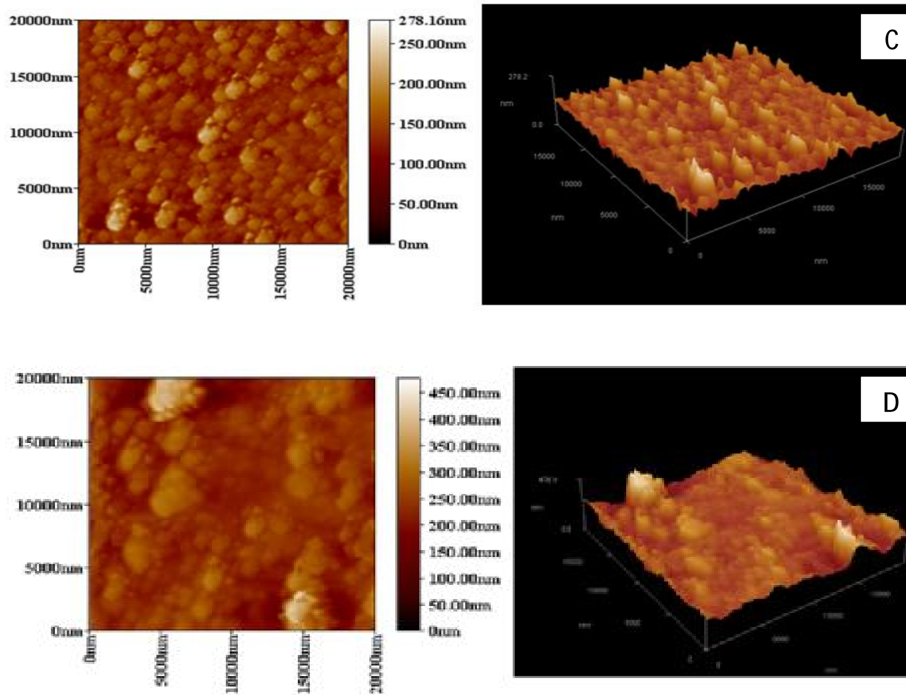


Figure (2) AFM surface morphologies of copper oxide thin films.

Figure(3) shows the effect of discharge current on the thickness of the deposited films, because the increasing of discharge current leads to increase in the density of charge particles included the bombarding ion of argon which influenced in yield rate of sputtering from the Cu target.

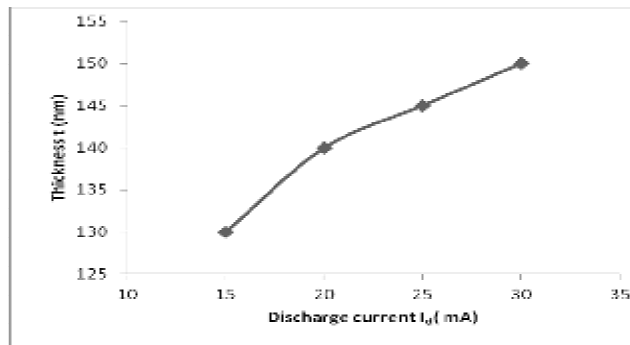


Figure (3) The Variation of film thickness as a function of the discharge current.

Figure (4 and 5) show the optical absorbance and transmittance spectrum of deposited films as deposited at different thickness, which are prepared at various glow discharge currents in the wave length range of 300 to 1100 nm at room temperature ,which increases and decreases respectively with increasing of film thickness. This effect could be attributed to increase the diffusion of copper and oxygen atoms into band gap as localized levels.

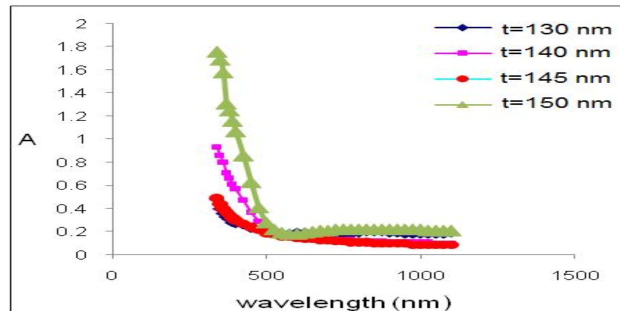


Figure (4) Absorption spectra of copper oxide films deposited with different thicknesses.

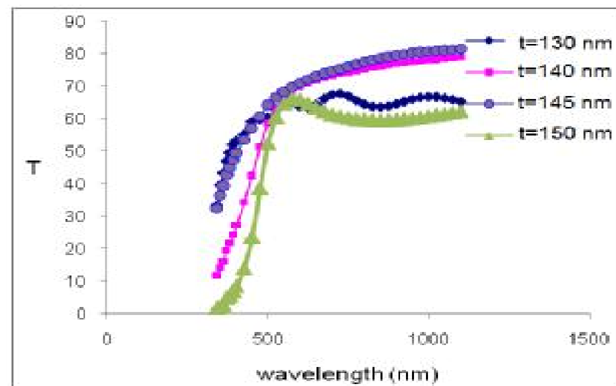


Figure (5) Transmission spectra of copper oxide films Deposited with different thicknesses.

The direct energy gap of prepared films has been determined from the plot of $(\alpha h\upsilon)^{2/3}$ the absorption coefficient as a function of photon energy as shown in Figure (6). The value of the direct energy gap was found to be (2.3, 2.2, 2.1, 2.05) eV for deposited films this result agreement with studying of Mugwang 'n *et al* [13] . This slight reduction in direct energy gap is due to the copper oxygen diffusion which occupies interestial essential sites between Cu_2O lattices. This process results in new localized states of impurities, which causes decreases in the optical energy gap. The behavior of refractive index and extinction coefficient are similar to the corresponding transmittance and absorbance spectra respectively.

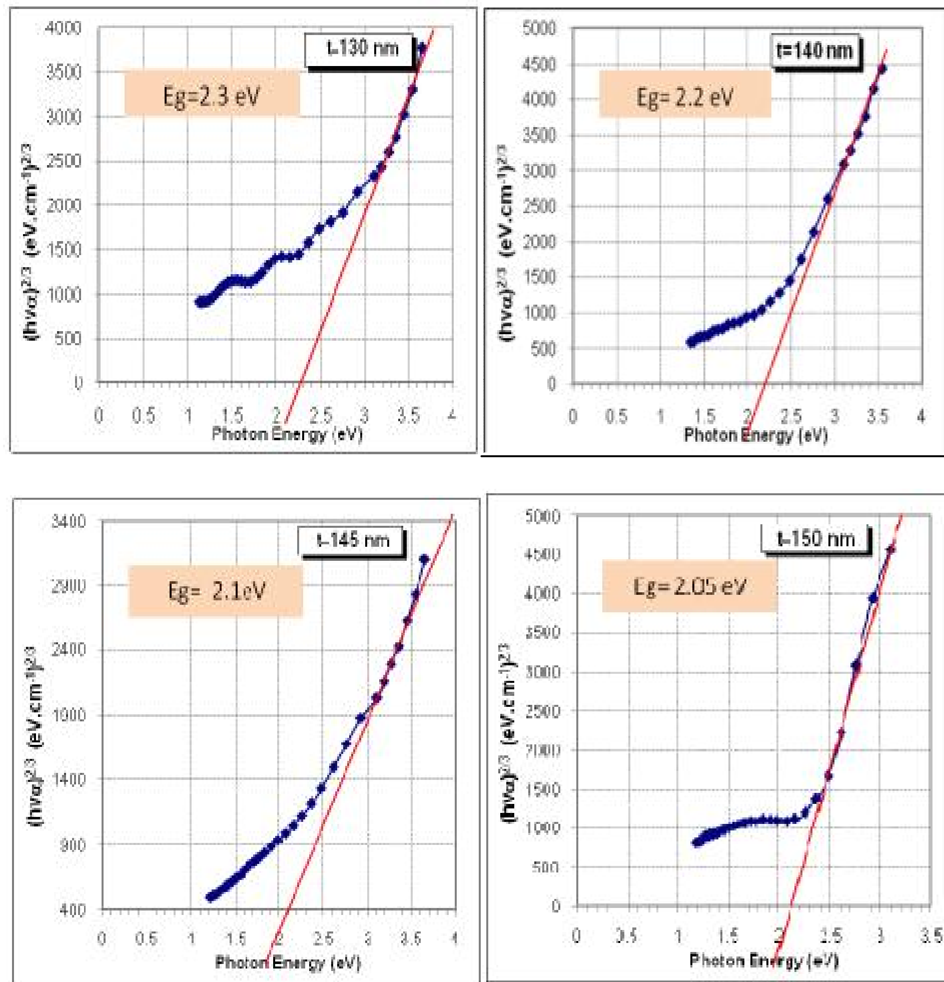


Figure (6) The variation of $(\alpha h\upsilon)^{2/3}$ with photon energy of Copper oxide thin films for different thicknesses.

CONCLUSIONS

We successfully obtained sputtered Copper Oxide thin films. For the prepared films, spectral investigation of the transmittance and absorbance at (300-1100) nm region showed that the optical properties of Cu_2O films are affected by films thickness which related to the sputtering discharge current.

REFERENCE

[1]. Roos, A. T.Chibuye, B.Karlsson, Solid Energy Mater.7, 453 (1983)
[2]. Matsuzaki, K. K.Nomura, H.Yanagi, T.Kamiya, M.Hirano, Appl.Phys.Lett 93, 202107(2008).
[3]. Scanlon,D.O. B.J.Morgan, G.W.Waston, Phy.Rev.Lett.1, 2582(2010).
[4]. Reddy, A.S. S.Uthanna, P.S.Reddy, Appl.Surf.Sci.253, 5287 (2007).
[5]. Sharma,S.P. J.Vac.Sci.Technol.16,1557 (1979).

[6].Ogwu, A.A. E.Bouquerel, O.Ademosu, S.Moh, E.Crossan, F.Placido,An investigation of the surface energy and optical transmittance of copper oxide thin .Ims prepared by reactive magnetron sputtering thin film center,Electronic Engineering and physics Division, Acta Materialia 53:5151-5159 (2005).

[7]. Samarasekara P.Yapa NU S.Kumara NTRN and Perera MVK Bull.Mater.Sci.30 (2) 113 (2007).

[8]. Samarasekara P.Mallika Arachchi M A K .Abeydeera A S.Fernando C an N.Disanayake a S and Rajapakes RMG Bull.Mater.Sci.28 (5), 483 (2005).

[9].YuZQ .Wang C M.Engelhrd M H.Nachimuthu P.Mc Cready D E.Lyubinetsky IV and Thevuthasan S Nanotechnology 18, 115601 (2007).

[10]. Seong Ho Jeong and Eray Shadily J.Crystal Growth 311 (17)4188 (2009).

[11]. Shogo Ishizuka, Shinya Kato, Takahiro Maruyama and Katsuhiro Akimoto Jpn.J.Appl.Phys.40 2765(2001).

[12].Bhattacharyya,S.R.D.Reppin,P.Sanguino,R.Ayonchi,A.Polity,R.Schwar,D.Hofmann,B.K.Meyer,Acta Phys.Polonica,120,11A (2011).

[13]. Mugwang'a F.K , Karimi P.K , Njoroge W.K , Omayio O2 and Waita S.M, Int. J. Thin Film Sci. Tec. 2 No. 1, 15-24 (2013).