IOSR Journal of Engineering (IOSRJEN) **www.iosrjen.org**
ISSN (e): 2250-3021, ISSN (p): 2278-8719
Vol. 08, Issue 3 (March. 2018), ||V2|| PP 82-87

Electron Beam Evaporated Copper Oxide Thin Films

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ABSTRACT: Cuprous oxide (Cu_2O) thin films were deposited on glass substrates by electron beam evaporation under various accelerating voltages. The films deposited at lower accelerating voltage exhibited slightly high oxygen content. The crystallite size of films was increases from 17 to 29nm with increasing the accelerating voltage from 2 to 4kV. The root mean square (RMS) surface roughness of the films was decreasing with increasing the accelerating voltage. As increasing the accelerating voltage, photoluminescence (PL) peak intensity increased and an additional peak was observed at 539nm (2.3eV). The films deposited at accelerating voltage of 4kV exhibited highest transmittance of 79% with electrical resistivity of 31Ωcm.

KEYWORDS: Accelerating Voltage, Cuprous Oxide, Electron Beam Evaporation, Thin Films

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Date of Submission: 26-02-2018 Date of acceptance: 05-03-2018

I. INTRODUCTION

Cuprous oxide (Cu_2O) is p-type semiconducting material with band gap of ~2.1eV, high absorption coefficient in the visible light region, nontoxicity, abundance and low fabrication cost[1-3]. Cu_2O has wide range of applications including solar cells, sensors, photocatalysts lithium ion batteries, transparent displays[4-7]. Several thin film deposition techniques such as chemical vapour deposition[8], thermal oxidation[9], sputtering[10] and electron beam evaporation[11] have been used to prepare the Cu_2O films. In the literature, very few reports were available on the electron beam evaporated Cu_2O films. Electron beam evaporation (EBE) is a versatile technique due to its unique properties such as uniformity of the films over the substrate, useful for depositing alloy and compound materials, films having the stoichiometry close to the bulk and material loss is minimal. The properties of the electron beam evaporated films are mainly depends on the deposition parameters such as accelerating voltage, substrate temperature, target to source distance, ambient pressure, thickness, etc. In the present work, Cu_2O films were deposited on the glass substrates at various accelerating voltages and studied the effect on the compositional, structural, surface morphology, electrical and optical properties of the films.

II. EXPERIMENTAL

The Cu_2O thin films were prepared on the glass substrates using high purity Cu_2O pellets by electron beam evaporation. The vacuum chamber was pumped with the combination of diffusion pump and rotary pump and is capable of creating an ultimate vacuum of $4x10^{-4}$ Pa. The pressure was measured using combination of Pirani–Penning gauge. The pellet was prepared using high purity (99.99%) Cu_2O powder. The pellets were kept in a water-cooled copper crucible. The films deposited at different accelerating voltages by keeping the other parameters at constant values. The deposition parameters maintained during the preparation of Cu_2O films are given in Table 1.

The chemical composition of the films was analyzed by Energy Dispersive Spectroscopy (EDS) attached with SEM of model Oxford instruments Inca Penta FET X3. The crystallographic structure of the films was analyzed by Seifert 3003TT X-ray diffractometer (XRD), using Cu Kα radiation (k = 0.1546 nm). The microstructure and surface morphology of the films was studied by scanning electron microscopy (SEM) and atomic force microscopy (AFM), respectively. The photoluminescence (PL) spectra were measured with a LS 55 fluorescence spectrometer (Perkin Elmer) at room temperature. The electrical properties of the films were

measured by using standard four-probe method. The optical transmittance of the films was recorded using a UV–Vis–NIR double beam spectrophotometry.

Table.1. Deposition parameters of Cu_2O films during deposition

Deposition method		: electron beam evaporation
Power source		: e-beam power supply (3kW)
Pellet		: Cu_2O (10mm dia and 3mm thick)
Substrates		: Glass
Target to substrate distance		: 60mm
Ultimate pressure	(P_U)	: $4x10^{-4}$ Pa
Evaporation pressure	(P_W)	: $3x10^{-2}$ Pa
Substrate temperature	(T_S)	: 473 K
Accelerating voltage		: 2, 4 and 6kV
Filament current		: 30mA
Deposition time		: 5 to 16min
Films thickness		: 192nm

III. RESULTS AND DISCUSSION

Fig.1. shows the EDS spectra of Cu_2O films at various accelerating voltages. The accelerating voltage slightly influenced the composition of the films, and no reflections of impurity were detected. From the EDS results, films have high oxygen content at lower accelerating voltage and the films deposited at accelerating voltage of 4kV were near to the stoichiometry. The deviation of the films composition could be due to atomic segregations occurs in the target during the electron beam and mater interaction or presence of the gas in the ablation chamber[12]. The obtained composition results at different accelerating voltages were listed in Table 2.

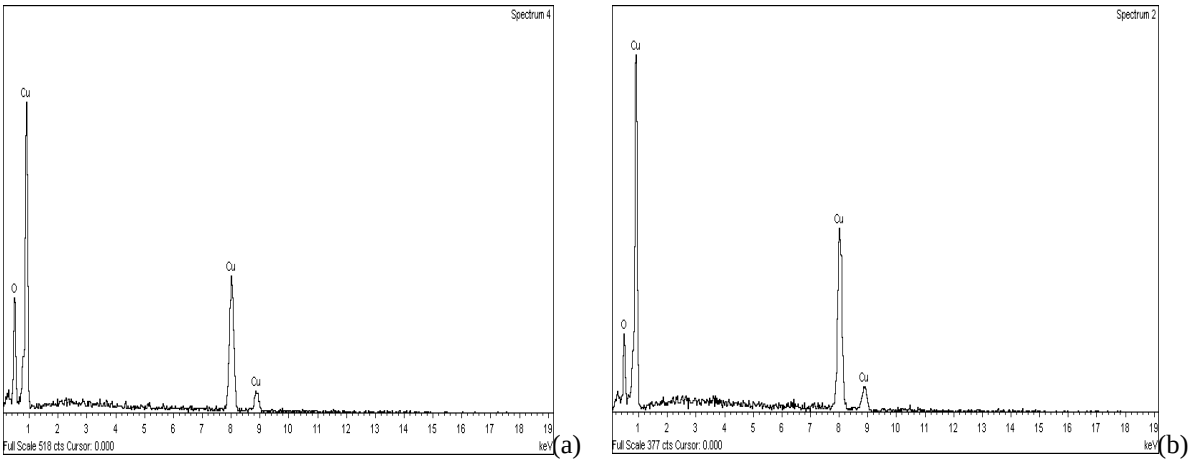

Fig.1. EDS spectra of Cu_2O films at different accelerating voltages: (a) 2kV and (b) 4kV

Table 2: The compositional results of Cu_2O films at different accelerating voltages

Accelerating voltage	Element	Atomic percentage
2 kV	O K	38.83
	Cu K	61.17
4 kV	O K	35.39
	Cu K	64.61

Structural properties

Fig.2. shows the XRD patterns of Cu_2O films deposited at different accelerating voltages. The films exhibited Cu_2O phase only and grown preferentially on (111) orientation. The crystallinity of the films increases and an additional peak of (220) appeared with increasing of accelerating voltages to 4kV. Beyond this accelerating voltage the crystallinity of the films decreased and additional peak (220) was disappeared. The penetration depth of the electrons in the target is influenced by the accelerating voltage. As increasing the accelerating voltage the electrons become more energetic and travel with higher velocity and increase the

penetration depth. Consequently, the energy of the ablation species striking the growing films is higher and which are influenced the crystallization process of the films[13]. The accelerating voltage must be sufficiently low to have adequate nucleation and growth.

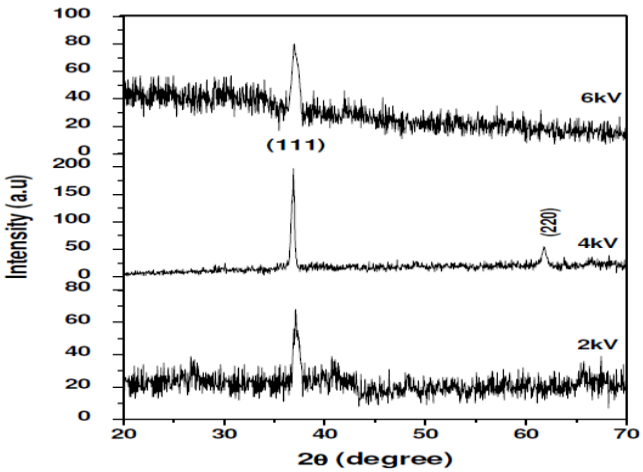

Fig.2. XRD patterns of Cu_2O films at different accelerating voltages

The average crystallite size of the Cu_2O films at various accelerating voltages was calculated by using Scherrer's equation[14]. The crystallite size of films was increases from 17 to 29nm with increasing the accelerating voltage from 2 to 4kV, and it decreased to 14nm at higher accelerating voltage of 6kV. The kinetic energy of ablation species emitted from the target is sufficiently high and it gives high surface mobility to these species as increasing the accelerating voltage. Consequently the structural defects are reduced and crystallinity of the films increased. However, the ablation species with too high kinetic energy may damage the crystallization of the films.

The lattice parameter (a) of the films was calculated using the following relation,

$$d = a / (h^2+k^2+l^2)^{1/2} \qquad \text{----------(2)}$$

where h, k and l are the Miller indices. The interplaner spacing (d) was evaluated from the X-ray diffraction data using the Bragg's relation. The obtained lattice parameter values are lower than the standard value (ICDD = 4.269Å) and it decreased with increasing of the accelerating voltages. The obtained lattice parameter values are 4.237, 4.226 and 4.188 Å for accelerating voltages of 2, 4, and 6 kV, respectively.

Microstructure and surface morphology

Fig.3. shows the SEM images of Cu_2O films deposited at different accelerating voltages. The films exhibited fine grains at accelerating voltage 2kV and the grain size becomes bigger, uniform and smooth surface with increasing the accelerating voltage to 4kV due to decreasing of structural defects, and agglomeration of smaller grains. The films deposited at higher accelerating voltage exhibited fine grains and islands.

The surface roughness is an important aspect since it influences the electrical and optical properties of the films. The AFM images of Cu_2O films at different accelerating voltages are shown in Fig.4. The root mean square (RMS) surface roughness of the films is 4.5, 2.8 and 3.8 nm for accelerating voltage of 2, 4 and 6kV, respectively. The decreasing of the surface roughness with increasing the accelerating voltage is due to increasing of the grains size of the films.

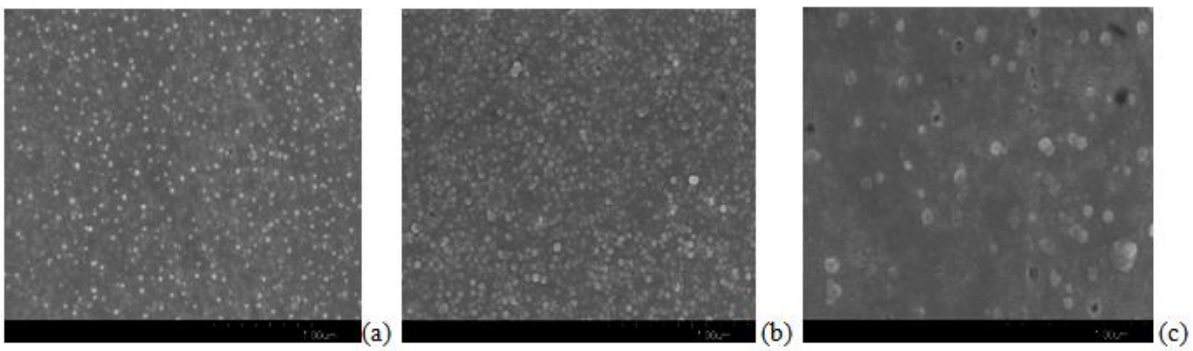

Fig.3. SEM images of Cu_2O films at different accelerating voltages: (a) 2kV, (b) 4kV and (c) 6kV

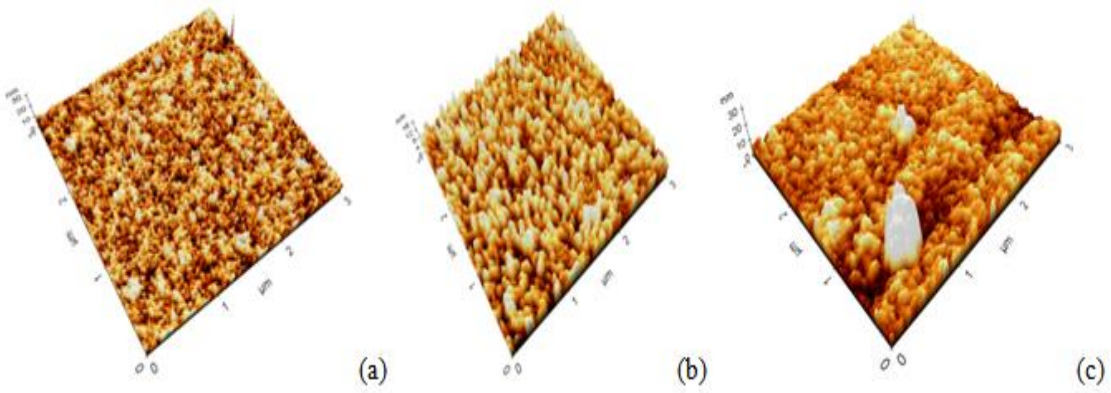

Fig.4. AFM images of Cu_2O films at different accelerating voltages: (a) 2kV, (b) 4kV and (c) 6kV

Photoluminescence properties

The Photoluminescence (PL) spectra of the sample determine that the produced material has enough quality or not[15]. Fig.5. shows the PL spectra of Cu_2O films at different accelerating voltages. The films deposited at lower accelerating voltage of 2kV shows emission peak with weak intensity at 500nm (2.48eV) for Cu_2O. As increasing the accelerating voltage, peak intensity increased and an additional peak was observed at 539nm (2.3eV). On further increasing the accelerating voltage the peaks intensity was decreased. The improvement of the emission peak intensity with accelerating voltage can be attributed to the decreasing of the structural defects.

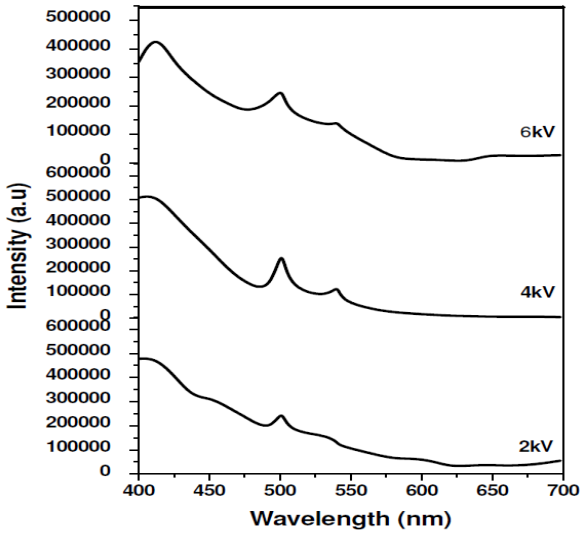

Fig.5. PL spectra of Cu_2O films at different accelerating voltages

Electrical and Optical properties

The electrical properties of Cu_2O films at different accelerating voltages are listed in Table 3. As the accelerating voltage increases the resistivity and carrier concentration of the films decreases and Hall mobility increases. This behavior could be associated to changes of the films composition and structural properties. The decrease of Hall mobility at higher accelerating voltage was may be due to lattice damage caused by very high energy ablation species. Fortunato et al.[16] observed the increasing of the electrical resistivity and decreasing of Hall mobility with increasing of the rf power in magnetron sputtered IZO films.

It is known that the optical properties have strong correlation with the structural and surface morphology such as density of defects, crystallinity and surface roughness of the films. Fig.6. shows the optical transmittance spectra of Cu_2O films at different accelerating voltages. The optical transmittance of the films increases with increasing accelerating voltage from 2 to 4 kV and decreases at higher accelerating voltage of 6kV. The increasing of the optical transmittance with accelerating voltage was due to increasing of the grain size and surface smoothness of the films.

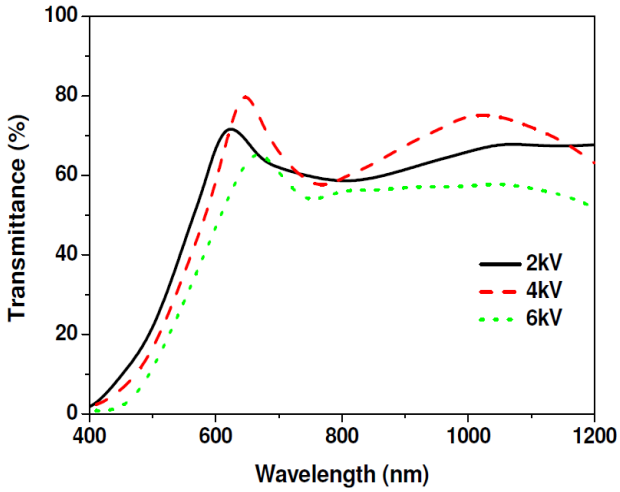

Fig.6. Optical transmittance spectra of Cu_2O films at different accelerating voltages

The optical band gap (E_g) of the films was evaluated from the extrapolation of the linear portion of the plots of $(\alpha h\nu)^2$ versus $(h\nu)$ (α is the absorption coefficient, $h\nu$ is the photon energy). The optical band gap values of Cu_2O films as a function of accelerating voltage are listed in Table 3. The band gap decreases with increasing of accelerating voltage. The decreasing of band gap values with increases of accelerating voltage is due to reduction of structural disordering and changes in microstructural properties.

Table 3. Electrical and optical properties of Cu_2O films at different accelerating voltages.

Accelerating voltage	Resistivity (Ωcm)	Hall mobility (cm^2/V.sec)	Carrier concentration (cm^{-3})	Transmittance (%)at λ=650 nm	Band gap (eV)
2kV	74	3.2	$4.2x10^{16}$	73	2.31
4kV	31	7.2	$2.8x10^{16}$	79	2.20
6kV	22	4.9	$5.7x10^{16}$	69	2.13

IV. CONCLUSION

Cu_2O films of have been deposited on glass substrates by electron beam evaporation. The films deposited at accelerating voltage of 4kV were near to the stoichiometry. From XRD results, the films exhibited only Cu_2O phase, no impurities such as Cu, CuO was observed. The films deposited at accelerating voltage of 4kV exhibited better crystallinity and smooth surface. The electrical resistivity of the films decreased and optical transmittance increased with increasing the accelerating voltage. The present obtained results make Cu_2O film is a promising candidate p-type semiconductor material for solar cell applications.

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V. Sravanthi " Electron Beam Evaporated Copper Oxide Thin Films." IOSR Journal of Engineering (IOSRJEN), vol. 8, no. 3, 2018, pp. 82-87.