ISSN: 0976-3376

Available Online at http://www.journalajst.com

ASIAN JOURNAL OF SCIENCE AND TECHNOLOGY

Asian Journal of Science and Technology
Vol. 07, Issue, 09, pp.3574-3576, September, 2016

RESEARCH ARTICLE

STRUCTURAL, OPTICAL AND ELECTRICAL PROPERTIES OF CuO THINFILMS PREPARED BY SPRAY PYROLYSIS

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ARTICLE INFO

Article History:
Received 14th June, 2016
Received in revised form
29th July, 2016
Accepted 19th August, 2016
Published online 30th September, 2016

Key words:

CuO thin films,
Spray pyrolysis,
Bandgap,
Hall coefficient.

ABSTRACT

Using the spray pyrolysis technique to prepare the P-type CuO thin films. It is carried out for one molar concentrations of 0.1M at 250°C from copper acetate precursor solution on the glass substrate. The study of X-ray diffraction attained all the films exhibit polycrystalline nature with monoclinic crystal structure comprised uniformly distributed grains. The electrical properties of the films like mobility, conductivity Hall co-efficient (RH), and carrier concentration have been studied. Hall Effect measurement is to confirm the p-type conductivity of the films. The bandgap energy size, resistivity and average particle were also determined. The resistivity has been investigated by four probe method for different molar concentrations of copper oxide.

INTRODUCTION

Different transition metal oxides (copper, iron, nickel, zinc and cobalt) have numerous applications. One of these is copper oxide (CuO) as an important P-type transition metal semiconductor oxide has been extensively studied. CuO has been established as a number of applications like gas sensors (Mariammal *et al.,* 2013; Hübner *et al.,* 2011; Manish Kumar Verma and Vinay Gupta, 2012), solar photovoltaic (Chandrasekaran, 2013; Amun *et al.,* 2013), lithium ion electrode (Morales *et al.,* 2005) etc. There are various established ways of fabricating CuO thin films like spray pyrolysis (Morales *et al.,* 2005; Madhav Singh *et al.,* 2008; Nasse *et al.,* 1998; Iqbal Singh and Bedi, 2011), spin coating (Hong Youl Bae, and Gyeong Man Choi, 1999; Aykut Nalbant *et al.,* 2013), dip coating (Zhang *et al.,* 2013; Gulen *et al.,* 2013), SILAR (Mageshwari and Sathyamoorthy, 2013; De Los Santos Valladares *et al.,* 2012) to name a few. Among all these spray pyrolysis technique has stoichiometry in multi-component system and splendid control of chemical uniformity.

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As the presence of acceptor levels attributed to copper vacancies, CuO is considered to be a p-type transition metal semiconductor with a narrow band gap of 2.34 eV (De Los Santos Valladares *et al.,* 2012), and therefore it may be effective to construct PN junction diodes. In pure condition, the stoichiometry CuO material is an electrical insulator. However, CuO is subjected to a chemical spray, it gets deviated and thereby causes from stoichiometry due to defects and impurities.

Experimental

In this study, the copper acetate dissolving in distilled water to deposit CuO thin films was constructed by spray pyrolysis method. The resulting solution was mixed for 1 hours using a precursor solution. The final solution, with concentration of 0.1 M was dark blue and clear, without any suspension of particles. The solution was sprayed onto the ultrasonically cleaned glass substrates. The substrate temperature was maintained at 250oC and it was measured using thermocouple. The solution flow rate was controlled by a flow meter and kept at 2 ml min−1and the distance between the nozzle and the substrate was maintained at 18cm. The thickness of the films was measured using gravimetric method. The crystalline

structure of the cop peroxide films was studied by X Pert Pro X-ray diffract meter (XRD), with CuKα radiation. The Ecopia HMS-3000 version 3.51.3 Hall effect measurement system was used to measure the Hall-coefficient, charge carrier concentration, mobility, resistivity and conductivity of the films.

RESULTS AND DISCUSSION

Characterization by X-rays

Fig.1 shows X-ray diffraction patterns are presented for CuO thin films deposited at 250o C for 0.1 molar concentration. The obtained XRD pattern of the films for 0.1 molar concentration unveiled the CuO with single phase monoclinic structure. The peaks at 2θ angle 35.15and 38.30, with dhkl 2.55 Å and 2.34 Å, correspond to (-111) and (111) planes respectively. (JCPDS 03-1018). From the xrd reports, increasing the precursor concentration increases the film thickness results the rise in peak intensity during the film formation. At 0.1 M concentration peaks at 35.56 and 38.65 appeared, due to (111), (200) planes respectively.

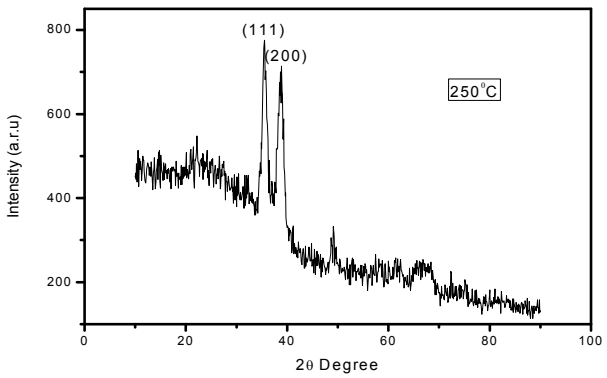

Fig.1. XRD Patterns of CuO thin film for molar concentrations

All the peaks in the XRD pattern represent the monoclinic structure of CuO. The crystallite size (D) was calculated using Scherrer's formula (Gopalakrishna *et al.,* 2013).

$D = 0.9\lambda/\beta \cos\theta$

Where D is the crystalline size, β is the broadening of diffraction line measured at half of its maximum intensity and λ is the X ray wavelength. Fig 2. SEM images were recorded to examine the surface morphology of the as deposited CuO thin films and the images are shown in Fig. The as deposited films have islands of different sizes and shapes, and their distribution on the surface is not homogeneous. These could be the result of the chemical reaction during the deposition. SEM micrographs reveal the formation of particles with different shapes and size, it seems appropriate to consider that the particle which appear in SEM images are, in fact, grain agglomerates. From table.1 the crystalline size observed that increasing the concentration.

The optical absorption spectra were recorded in the wavelength region 300 – 900 nm and the band gap was determined from (hν)1/2 against hν graph (Fig.3) Band gap observed as 2.34 eV for the molar concentration for 0.1 at % respectively.

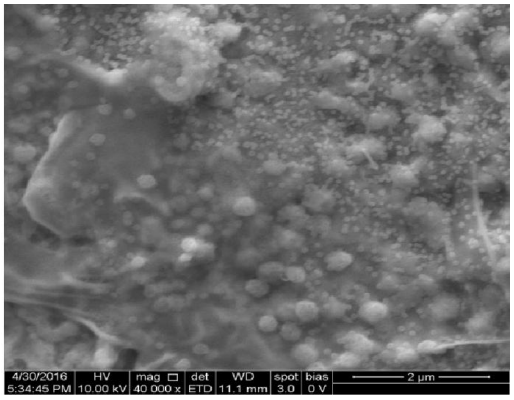

Fig. 2. SEM image of CuO thin films at 250°C

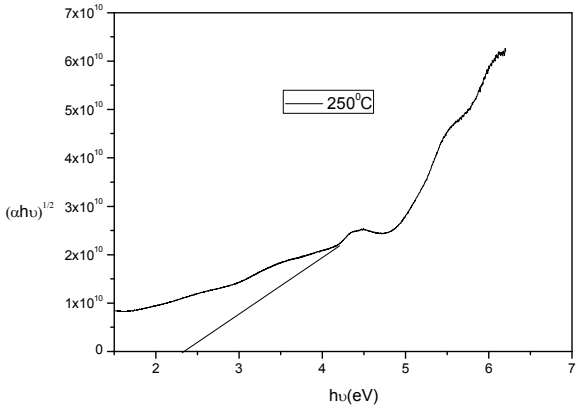

Fig. 3. Optical Bandgap Energy

Fig.4. shows that the variation of k with energy, the k value for all the films behaves a linear trend upto 2 eVafter that there is a gradual decrease in the extinction coefficient value. The blue shift in the extinction coefficient value denotes that the films are stronger absorbing medium in this range.

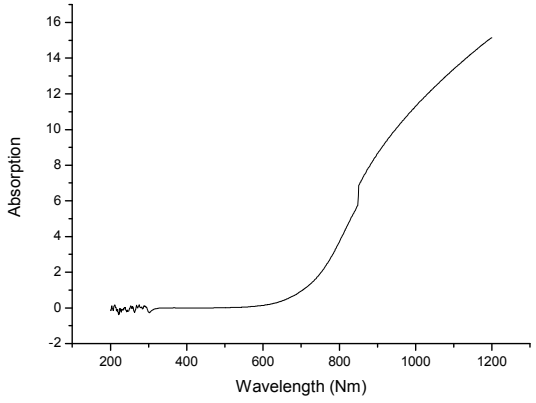

Fig.4. Absorption co-efficient

Table 1. Electrical properties

Sample	E_g (eV)	$R_H 10^7$ (cm^3/C)	$\Sigma 10^{-4}$ ($\Omega^{-1}.cm^{-1}$)	M 10^3 (cm^2/Vs)	P 10^4 (Ωcm)	N 10^{14} (/cm^3)	D nm
0.1 at%	2.34	0.50	4.3	2.46	0.15	3.5	4.2

The another important optical constant, extinction co-efficient (k) has been calculated the following formula[17]

$k = \alpha\lambda/4\pi$

Electrical properties

The Hall Effect measurements of the CuO thin film deposited with different molar concentration show p type conductivity and Hall coefficient increases with increasing solution concentration. The resistivity at lower concentration was first observed to be 0.18x104Ω.cm. The molar concentration increases, the resistivity increases due to decrease in the carrier concentration.

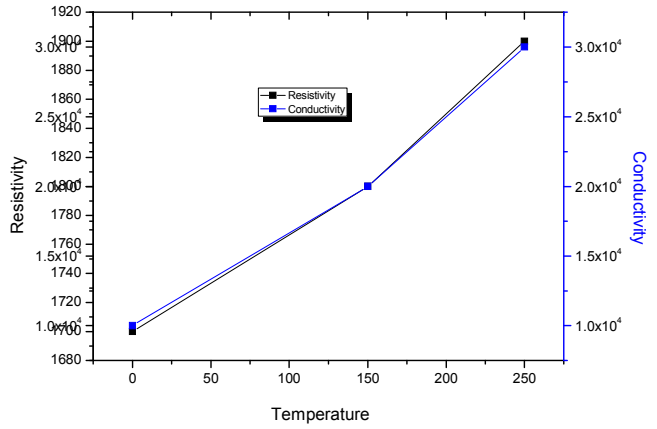

Fig. 5. Resistivity and Conductivity

Conclusion

The consequence of variation in the precursor concentration on the optical and electrical properties and SEM images of was discussed for CuO films deposited at 250°C by spray pyrolysis technique. The crystallite size was found from 4.2 nm with increasing in concentration. The highest figure of merit occurred for the film grown at 250°C with an optical transmittance about 76% in the wavelength range of 300 – 900nm. The results suggest that high-quality CuO thin film can be produced when capacity deposited growth at a temperature of 250°C. The obtained experimental results indicate the suitability of this material as transparent and conducting window materials in thin film solar cells and good for semiconducting materials.

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