Effect of low level nozzle height on properties of copper oxide absorption layer prepared by fully computerized spray pyrolysis depositions

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Abstract

The effect of approaching nozzle jet from the deposition surface on structural, optical and morphology properties of copper oxide thin films was studied. The film was prepared by homemade fully computerized CNC spray pyrolysis deposition technique at preparations speed (3, 4, 5, and 6 mm/sec). The repeated line mode was used at deposition temperature equal 450 °C whereas the spraying time was in the range of (15-30 min) according to the deposition speed. The film exhibit polycrystalline structure with preferred orientation along (-111), (022) and (011), (002) at a 2θ value of (35.63^o) and (38.8^o) respectively. Optical band gaps were recorded at these speed shows variance in value from (1.53-2.08 eV). Films thickness were found to be in the range (128-412 nm) which depends on preparation speed.

Key words

Fully computerized Spray pyrolysis, Preparation speed, X-ray diffraction, Optical properties.

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تأثير اقتراب مستوي ارتفاع الفوهة على خصائص طبقة الامتصاص من اوكسيد النحاس المحضرة بواسطة منظومة الترسيب بالرش الحراري المتكاملة والمحوسبة

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الخلاصة

تم دراسة تأثير قرب المسافة بين جهاز التذرية من سطح الترسيب على الخصائص التركيبية، والبصرية، والتجانس لأغشية أوكسيد النحاس. تم تحضير الاغشية بواسطة منظومة الرش الكيميائي الحراري المتكاملة والمحوسبة بواسطة السيطرة العددية للكومبيوتر المصنعة محليا عند سرع تحضير (3، 4، 5 و 6) ملم/ثا. استخدم نمط الطلاء المتكرر عند درجة حرارة التحضير 450 درجه مئوية حيث كان زمن الرش في المدى (15-30) دقيقه تبعا لسرعة الترسيب المستخدمة. الأغشية أظهرت تركيب بلوري متعدد عند اتجاهات مميزه كانت (-111)، (022) و (011)، (002) عند القيمة زاوية (35,63) و (38,8) تباعا. سجلت قيمة فجوة الطاقة البصرية عند سرع الترسيب المستخدمة وكانت قيمها بداء من (1.53-2.08). سمك الأغشية وجدت في المدى (128-412) نانومتر اعتمادا على سرع الترسيب المستخدمة.

Introduction

Different chemical depositions were used to prepare copper oxide (CuO) thin films. Spray pyrolysis deposition (SPD) was one of them. Because of their optical properties such as high absorption coefficient in visible range[1] and energy band gap [2–4], make it important in solar energy application [5–7]. Furthermore, CuO have been prepared by using different techniques such as RF Plasma sputtering [8, 9], Radio Frequency(RF) Magnetron Sputtering [10,11], dc Magnetron Sputtering [12], active reactive evaporation [13], thermally oxidation [14] chemical and chemical vapor deposition [15, 16], Sol-gel [17], chemical spray[18,19]. CuO absorption layer can be used in wide range of application based on their properties and the examples include, the Magnetic behavior of copper oxide have been reported [20], CuO has been used as anode material of lithium-ion battery [5-6]. CuO regard as an interesting material used in environmental catalysis [22–25]. Although CuO vastly used as a different Gas sensor [26–28].

Experimental work

Copper oxide (CuO) layer were deposited by homemade fully computerized CNC spray pyrolysis Deposition (system design, mechanical, electrical, electronic design, controlling program C languages, graphical user interface GUI, under publishing) onto glass substrates from aqueous solutions of copper chloride, dissolved in distilled water, with fixed molar concentration equal to 0.1 M. compressed Air was used as the carrier gas with a constant flow rate equal 4.5 kg/cm^2. The aqueous solution of copper chloride was sprayed in atmospheric presser onto substrate area (10×10 cm^2) which is equal to four microscopic slides in the dimension (75×25×1.2 mm^3) and substrate temperatures 450°C and variance ± 5 at different preparation speed (3, 4, 5, and 6) mm/sec. While the nozzle – to – substrate distance was kept at the height 20 cm. Films thickness was measured by TFC Thin Film Measurement type (Black CR-25 USA) with resolution 5A^o and thicknesses measurement range (50 A^o – 20 μm). The structural properties of these films were characterized by X-ray diffraction using (Shimadzu XRD 6000). the diffraction pattern recorded with continuous scan mode and scanning speed 5deg/min and scan step of 0.1 and Cu Kα radiation (Wavelength (λ) =1.54060A^o). All samples were scanned in the 2θ range of (25-70) deg. The XRD spectra reveal that all obtained films sprayed at substrate temperatures equal to 450 °C have a poly-amorphous with monoclinic structure compared with (JCPDS File №. 05–0661), with preferred orientation along (-111) (002) and (111) (200) plans located at 2θ value of (35.63015^o) and (38.80148^o) respectively. Optical properties were monitored by using UV-Visible spectrophotometer type UV Mate SP8001. Transmittances spectra of the CuO layer measured in the range of 300–1100 nm at room temperature and the energy band gap was recorded.

Results and discussion

A. Structural properties

CuO layer which are prepared by using fully computerized spray pyrolysis technique (CNC SPD) techniques in different deposition speed, and deposited on glass substrate at 450°C, shows enhancement in the crystal structure in higher preparations speed as shown in Fig. 1a, 1b. This is due to density of droplet per unit area deposited on the substrate surface and this lead to the net of thermal energy quite enough to reach the hot substrate surface and results in subsequent decomposition/oxidation, and the opposite behavior occur in lower speed (3, 4 mm/sec). The thermal energy is not sufficient to got decomposition process in correct chemical vapor deposition (CVD). Crystallographic parameter for CuO layer at optimum deposition speed are listed in Table 1. Grain size of the films at different deposition speed was calculated using the Debye- Scherrer formula, and compared with (JCPDS) standard card № (05–0661) and present in Table 1 These results (2θ, lattice parameter d-spacing grain size) are consistent with standard card which previously reported. Fig. 1a, 1b show to as an improvement in the crystal structure, and this was approaching to the practical value (observed value of d, 2θ) from theoretical values form (JCPDS) card.

Table 1: Lattic parameter of CuO layer compared with standared card.

Depos- speed mm/sec	**2θ**	***I/Io***	***Avg. Grain size (nm)***	***d-spacing std°A***	***d-spacing obs°A***
5	35.598	69	21.34	2.523	2.51993
	38.78	100		2.323	2.3202
	58.237	14		1.581	1.58296
6	35.671	100	23.63	2.523	2.51934
	38.755	51		2.323	2.32185
	32.4843	12		2.751	2.75405

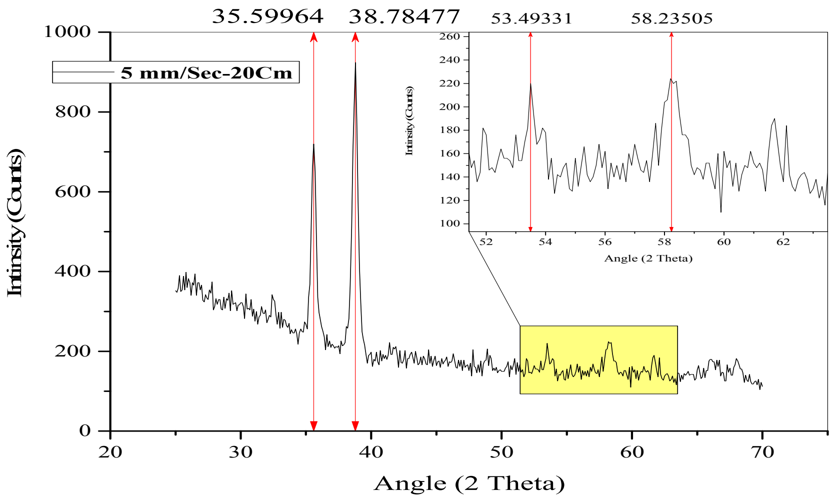


Fig. 1a: XRD pattren of CuO layer prepared at 5 mm/sec.

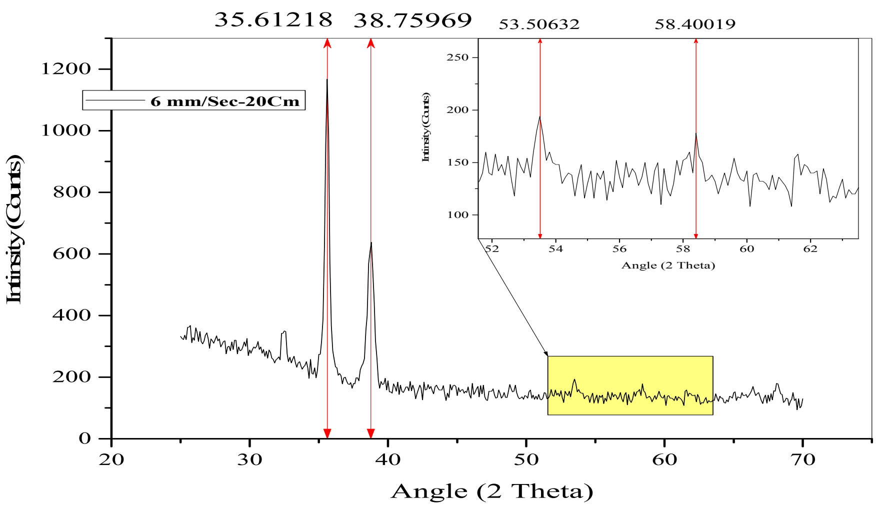


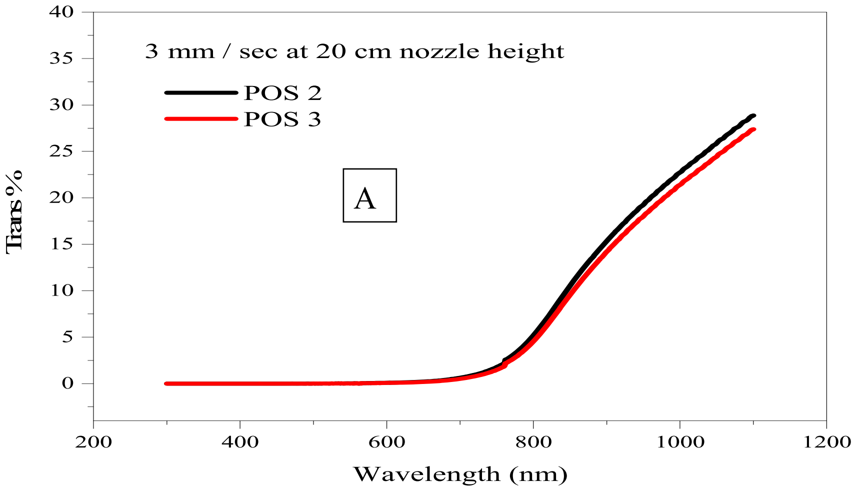
Fig. 1b: XRD pattren of CuO layer prepared at 6 mm/sec.

B. Optical properties

Transmittances spectra of the CuO layer were also measured. Fig. 2A, 2B, 2C, 2D shows the optical transmission T% as a function of the wavelength at two positions (POS) on the sample for the CuO layer at different deposition speed. All the films exhibit high absorbance in the studied UV–Vis range. The energy band gap, Eg, was calculated from the transmission spectra using the following relationship.

$$(\alpha h\upsilon)^2 = A\,(h\upsilon - E_g)^{\,n}$$

For the photon energy range shown in Fig. 3E, 3F, 3G, 3H, 3α reaches value higher than 10^4 cm^{-1}. This relatively high absorption coefficient is very important because the spectral dependence of α drastically affects the solar conversion efficiency. The inset shows the representation of $(\alpha h\nu)^2$ versus hν used for the calculation of the energy gap Eg. The films exhibit direct transitions corresponding to a band gap Eg in the range of (1.53-2.08) eV. Which is in good agreement with the solar application.



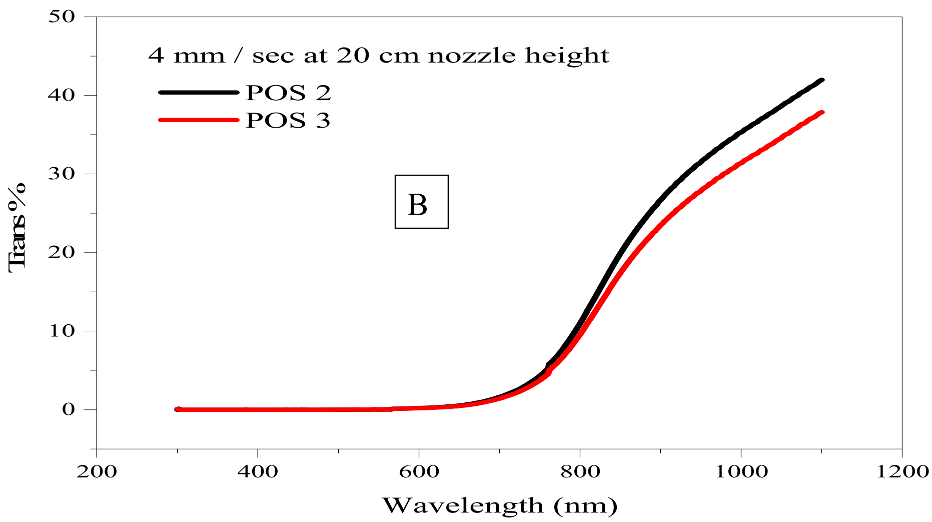
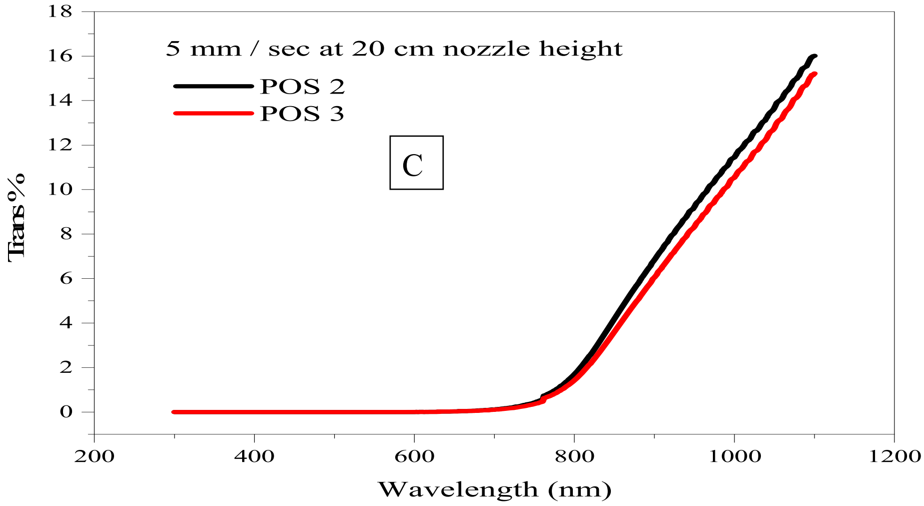


Fig. 2 (A, B): Tramsmitance spectra of CuO at different speed and postions (POS).



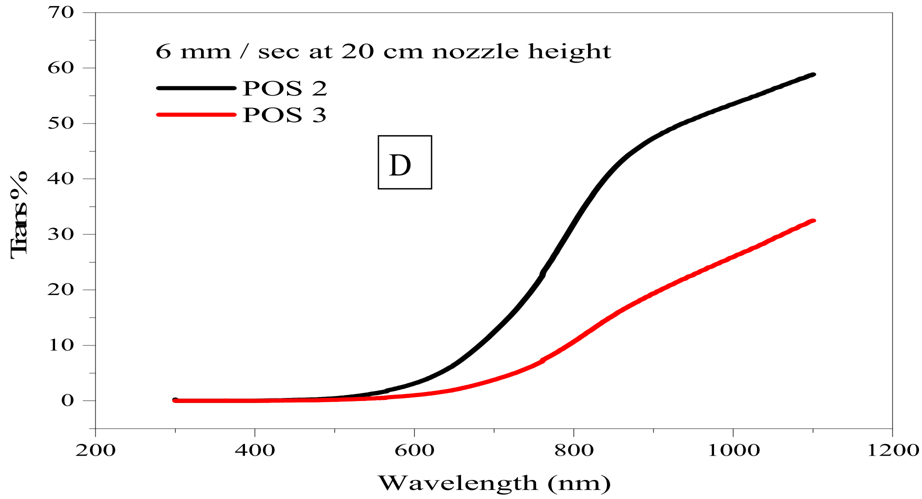


Fig. 3 (C, D): Tramsmitance spectra of CuO at different speed and postions(POS).

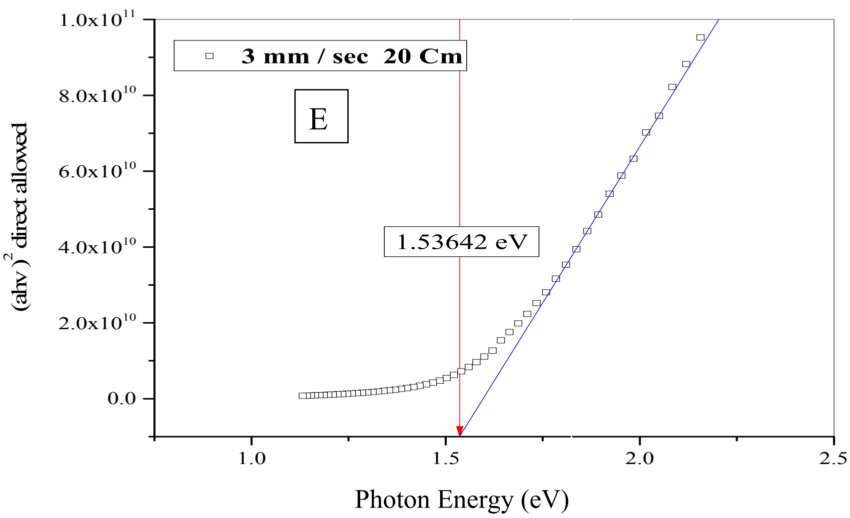


Fig. 4E: Energy band gap of CuO at different preparations speed.

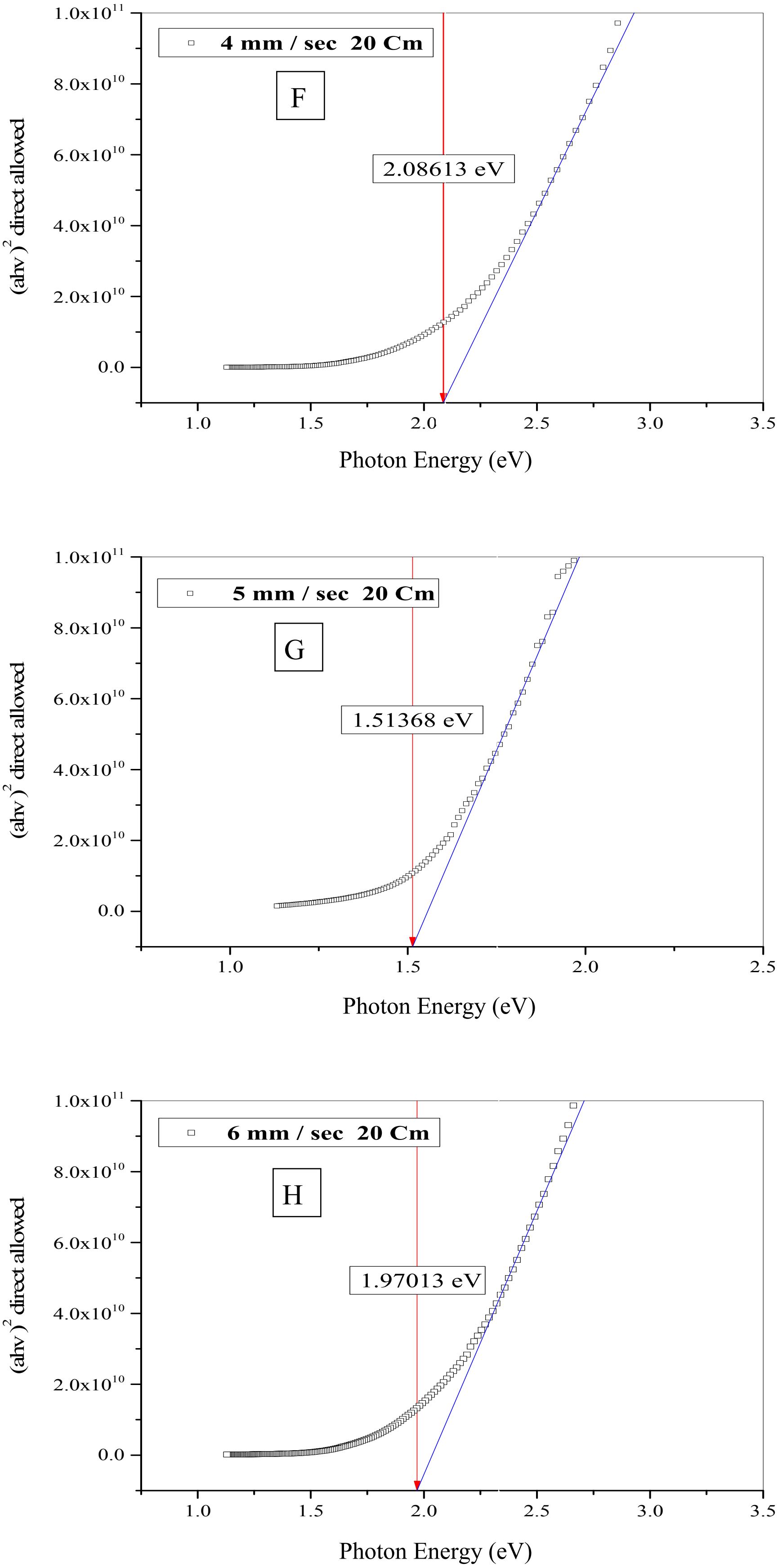


Fig. 5(F, G, H): Energy band gap of CuO at different preparations speed.

Conclosions

The spray deposition of CuO layer onto glass substrates at different preparation speed and nozzle-to-substrate distances equal 20 cm shows polycrystalline nature with monoclinic structure. The Transmittance spectra reveal that the transmittance decreasing with increased preparation speed. The higher speed exhibits an improvement on the structural and optical properties over preparation speed. The as- deposited film which changes to smooth and uniform at the speed (5, 6 mm/sec), band gap, thickness, found to be increased in odd number of speed than even number of speed. The band gap energy varies from 1.513 to 2.086 eV with change in preparation speed.

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