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ScienceDirect

Ceramics International 41 (2015) 4867–4873

CERAMICS INTERNATIONAL

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Influence of growth temperature on the physico-chemical properties of sprayed cadmium oxide thin films

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Received 17 October 2014; received in revised form 21 November 2014; accepted 8 December 2014
Available online 13 December 2014

Abstract

Transparent conducting cadmium oxide (CdO) thin films have been deposited onto the soda-lime glass (SLG) substrates using a facile and cost-effective spray pyrolysis (SPT) technique. The films have been deposited at various substrate temperatures ranging from 250 to 400 °C in steps of 50 °C. The influence of substrate temperature on structural, morphological and electro-optical properties of CdO thin films has been investigated. Thermo-gravimetric analysis (TGA) study indicates the formation of CdO by decomposition of cadmium acetate after 250 °C. The X-ray diffraction study reveals that all samples are polycrystalline with major reflex along the (111) and (200) planes, manifested with the homogeneous distribution of roughly spherical clusters all over the substrate of varying grain size.The optical study shows band gap ranging between 2.3 and 2.5 eV. The Hall effect measurement indicates that the resistivity decreases from 2.43×10^{-3} to 0.99×10^{-3} Ω cm while carrier concentration increases from 2.61×10^{20} to $5 \times 10^{20}/cm^3$ and mobility lies in the range of 8.26–12.55 cm^2/V s.

Keywords: Transparent conducting oxide; CdO; Spray pyrolysis technique (SPT); Carrier concentration; Thermo-gravimetric analysis (TGA)

1. Introduction

Transparent conducting oxides (TCOs) are essential for technologies that required both large-area electrical contact and optical access in the visible portion of the light spectrum. High transparency with useful electrical conductivity is achieved by selecting a wide-band-gap oxides like ZnO, TiO_2, IrO_2, SnO_2, Sn:In_2O_3, and CdO [1–6]. For optoelectronic applications, the transparent conductor must be carefully processed to maximize optical transmitivity in the visible regime, while achieving maximum electrical conductivity.

CdO has gained substantial interest due to its interesting electronic and optical properties such as carrier mobility, high conductivity with direct band gap energy making it suitable candidate for electro-optical applications [6].

Various deposition methods have been employed to prepare CdO thin films such as plasma laser deposition (PLD) [6], RF Sputtering [7], chemical bath deposition (CBD) [8], sol–gel [9], vacuum evaporation [10,11], metalorganic chemical vapor deposition (MOCVD) [12], successive ionic layer adsorption and reaction (SILAR) [13] and a spray pyrolysis technique (SPT) [5]. Among these methods, SPT is a most promising and widely used method to prepare variety of thin films such as metal oxides, superconducting materials and nanomaterials. It is a simple, cost-effective and non-vacuum method which has several advantages such as high purity, and easy control of chemical composition [5].

The substrate temperature is one of the key factors in deciding the physical, chemical and other properties of any thin film. In this work, we have focused on the optimization of spray-pyrolytic synthesis of nanocrystalline CdO thin films in terms of substrate temperature aiming for highly transparent conducting oxide films. The effect of substrate temperature on the various properties such

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http://dx.doi.org/10.1016/j.ceramint.2014.12.045
0272-8842/

as structural, morphological, electrical and optical properties has reported.

2. Experimental details

The CdO films were synthesized by using cadmium acetate [($Cd(CH_3COO)_2$] aqueous solution as a precursor. By using double distilled water, 0.1 M cadmium acetate [A. R. grade (99% pure), Loba Chemie Pvt. Ltd., Mumbai] was prepared and sprayed onto the preheated ultrasonically cleaned glass substrates. The deposition parameters like solution concentration (0.1 M), spray rate (4 ml/min), nozzle to substrate distance (33 cm), pressure of carrier gas (75 kg/cm^2) and quantity of spraying solution (20 ml) were kept constant. The optimized values are indicated in brackets. The substrate temperature was varied from 250 °C to 400 °C in steps of 50 °C using electronic temperature controller with an accuracy of ±5 °C. The films deposited at 250 °C, 300 °C, 350 °C and 400 °C were allowed to cool naturally at room temperature and were further used for their different characterizations. These films are denoted by T_{250}, T_{300}, T_{350} and T_{400}. All the as deposited films were faint yellowish-brown in color, transparent, uniform, well adherent to the substrates and pinhole free.

The deposited films were characterized by means of their chemical, structural, morphological, optical and electrical properties with the help of different characterization techniques. To select the range of substrate temperature for deposition, thermo-gravimetric analysis (TGA) of cadmium acetate was carried out using a TA instrument SDT Q600 V20.9 Build 20. The structural properties of sprayed films were studied by X-ray powder diffractometer Bruker AXS (D2-Phaser) using Cu K_α (λ=1.5418 Å) operated at 25 kV, 20 mA. The microstructural study was carried out using a JEOL JSM 6360 (Japan) scanning electron microscope. The 3 dimensional surface morphology of sample was studied using AFM images obtained from the INNOVA IB3BE model (Bruker, USA) in contact mode. The thickness of the samples measured using surface profiler make Ambios Technology model XP-1 (USA). The optical absorption spectrum was studied at room temperature within the 200–800 nm wavelength range using a spectrophotometer UV-1800 SHIMADZU. The room temperature electrical measurements were carried out with Hall effect set up in Van-der Pauw configuration.

3. Results and discussion

3.1. Thermogravimetric analysis

It is very important to determine the possible temperature range in which the metal oxide films are formed, hence thermo-gravimetric (TGA) analysis of cadmium acetate precursor powder, taken in the appropriate proportion has been carried out. The typical thermogram obtained for 4.029 mg cadmium acetate powder is shown in Fig. 1. The thermal evolution in air takes place in five consecutive stages with weight losses for which inflection point coincide with the temperature corresponding to exothermic and endothermic peaks in DTA trace. It is clearly seen that the loss of water from the precursor take place at various temperatures in the range of 25–140 °C, corresponding to which endothermic peaks are observed. The total weight loss corresponding to removal of both the physisorbed and chemisorbed water is calculated is about 13.97%. The rapid weight loss commences at about 250 °C, which is indication of onset of the thermal decomposition of the precursor. This regular weight loss continues up to 500 °C as shown in Fig. 1. The weight loss during this temperature is mainly due to the removal of acetate group from the precursor, which leads to the formation of cadmium oxide. Thus, this result indicates the formation of CdO by decomposition of cadmium acetate after 250 °C. The similar result has been reported by Deokate et al. [15].

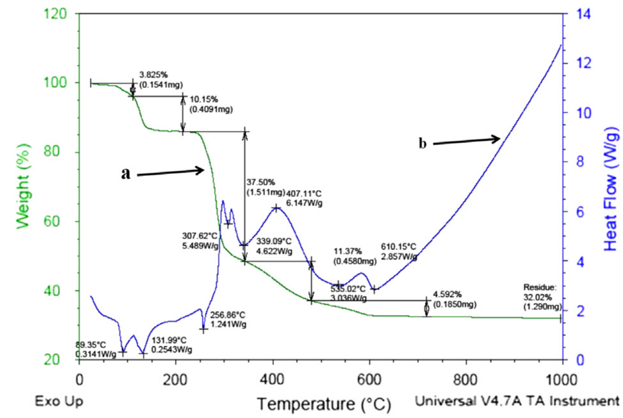

Fig. 1. (a) Thermo gravimetric analysis (TGA) and (b) differential thermal analysis (DTA) of the precursor powder of cadmium acetate in the temperature range 0–1000 °C.

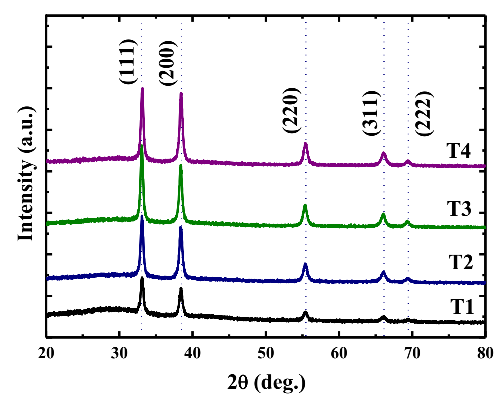

Fig. 2. The XRD patterns of CdO samples, T_{250}, T_{300}, T_{350} and T_{400}.

3.2. X-ray diffraction studies

Fig. 2 shows the XRD patterns of all the CdO thin films deposited at different substrate temperatures. It is found that all the films are polycrystalline in nature with face centered cubic (FCC) structure having major reflex along the (111) and (200) planes. The presence of other planes corresponding to (220), (311) and (222) has been observed. The observed 'd' values of the films are in well agreement with those reported for CdO