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Chemical Spray Pyrolysis Technique for Potassium Chloride Doped Cadmium Sulphide Deposition

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ABSTRACT

Three series of potassium cadmium sulphide (CdS) films were synthesized by spray pyrolysis method on glass substrates. The as-deposited films were structurally characterized using X-ray Diffractometry (XRD), Scanning Electron Microscope (SEM) and Energy Dispersive X-ray Spectroscopy (EDX) techniques. XRD results showed that the three samples are polycrystalline in nature exhibiting cubic structure a preferential growth texture (111). The average crystallite size determined by Debye Scherrer formula is in the range of 3.61-4.96 nm. SEM micrograph showed homogenous and uniform surface and EDX analysis confirms the presence of Cd and S atomic elements. From the optical characterization, the optical band gap energy for the as-grown thin films was found to be from 2.22 eV to 2.28 eV.

INTRODUCTION

Nowadays, doping chalcogenide materials by atomic elements has been subject of interest in materials science and thin films technologies [1-3]. This exercise is done in order to modify materials' properties for many applications. Chalcogenide materials doped can be used in diverse fields such as photovoltaic cells, electronic components, fabrication of large area photodiode arrays, photoconductors, sensors, antireflection coatings, optical filters, surface acoustic wave devices [2, 4], etc. Among the different metal sulphide chalcogenides, CdS is of great interest in scientific community owing to its remarkable properties [3].

Cadmium sulphide (CdS) is a wide direct band gap (E_g) of 2.42 eV, at room temperature. It is a n-type window layer widely used in thin film solar cells along with different absorbers such as CdTe, SnS, CdSe, Cu(In, Ga)Se_2 thin films [3]. From literature review, it is well known that one can control CdS properties by introducing a suitable dopant [4-8]. Several dopant atomic elements have been made to prepare CdS thin films including group I or III elements, transition metals, rare earth ions, etc. to improve the of CdS [1, 3, 4].

A number of methods have been developed for undoped and doped CdS films such as thermal vacuum evaporation method [5], Successive Ionic Layer Adsorption and Reaction (SILAR) [6], Chemical Bath Deposition (CBD) [7], spray pyrolysis technique [8], RF sputtering [9], Pulsed Laser Deposition (PLD) [10], etc. The simplicity, low cost equipment, easy coating of large surface deposition are some the advantages of spray pyrolysis method and hence it is adopted in the present work. To the best of our knowledge, a few reports available on potassium chloride-doping CdS have been studied. To this purpose, in this work, we have prepared K-doped CdS thin films using the spray pyrolysis technique. The influence of K concentration on morphological and structural properties of the as-grown CdS thin film was studied.

MATERIALS AND METHODS

All chemicals were of analytical grade (Merck) and we used them as received without further purification.

The starting solution is formed using an aqueous mixture of cadmium chloride ($CdCl_2$), 0.05 M, thiourea $(NH_2)_2CS$ 0.05 M and ammonium hydroxide NH_4OH. Then, to achieve

potassium doping, potassium chloride (KCl) with 1, 2 and 3 at.% concentrations were added to the starting solution to form the precursor solution which was sprayed on cleaned and heated substrates. The substrate temperature and the solution pH were maintained at 400 °C and 11 respectively. Three series of CdS:K thin films were prepared. Figure 1 shows the schematic diagram of spray pyrolysis method.

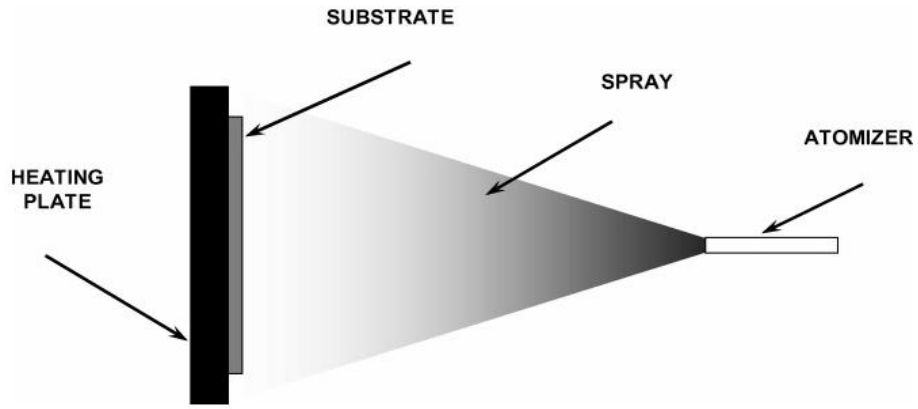


Figure No. 1: Schematic diagram of spray pyrolysis method

The morphology of the prepared samples were characterized using a scanning electron microscope (SEM) operating at 10 kV. EDX spectra were recorded using Model JEOL JSM 5600. The structure was determined by glancing angle X-ray diffraction (XRD, Philips X'pert MRD diffractometer) using Cu K radiation (λ =.1.5406 Å) at a scan rate of 4◦/min from 2θ~20° to 60°. A UV/Vis spectrophotometer was used to measure the reflectance (R) and transmittance (T) versus wavelength measurements.

The average crystallite sizes (D_{hkl}) of the samples were evaluated according to Debye Scherer equation [11]:

$$D_{hkl} = \frac{0.94\lambda}{\beta\cos\theta_{hkl}} \qquad (1)$$

Where λ represents the x-ray wavelength used for the measurement (λ = 1.5406 Å), β is the line width (FWHM) in radians, θ is the Bragg angle. Where λ is the X-ray wavelength (CuKα), θ is the Bragg diffraction angle, and β is the FWHM of the XRD peak appearing at the diffraction angle θ.

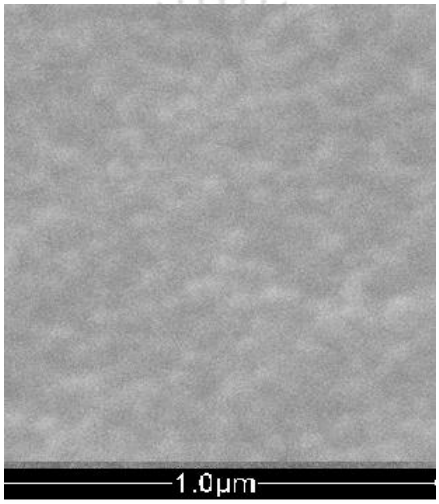
The direct energy band gap (E_g) was determined using the observed UV/Vis spectra and applied using Tauc's relation [12]:

$$(\alpha h\upsilon) = A\left(h\upsilon - E_g\right)^{\frac{1}{2}} \qquad (1)$$

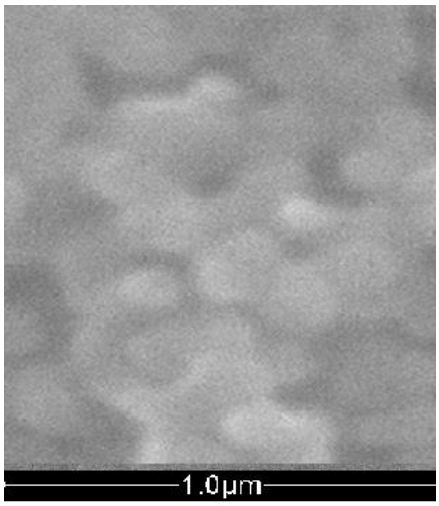
Where A is the constant that depends on the effective mass of the charge carriers in the material, α is the absorption coefficient, hυ is the photon energy.

RESULTS AND DISCUSSION

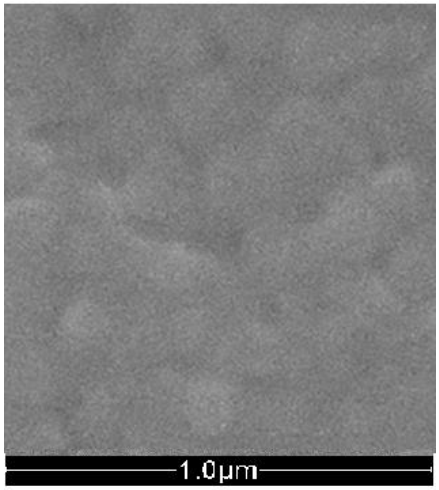
SEM observations are usually used to investigate the layer morphology. Figure 2 (a-c) shows the SEM images of K-doped CdS thin films prepared at 1 at.%, 2 at.% and 3 at.%. All the samples exhibit spherical nanoparticles which are uniformly distributed over the surface area. It is obvious that the crystalline structure of the three films are quite different. The difference is more clearly illustrated in the film surface. Figure 2a shows a cross section of the better film since the surface of this sample appears to be excellently smooth comparing to the others. Similar results have been reported by Fateh Mulla *et al.* [13].



(a)



(b)



(c)

Figure No. 2: SEM images of K-doped CdS thin films, deposited with 1 at.% K (a), 2 at.% K (b), and 3 at.% K (c)

A typical EDX spectrum of K doped CdS thin films, deposited with 1 at.% K is presented in Figure 3. We observe the presence Cd and S. Other impurities (Mg, Al, Ca) detected are from glass substrate The absence of potassium peak suggests that the addition of K^+ dopant to cadmium sulphide during the deposition does not create any change in the CdS matrix structure. This result matches well with previous reports [14]. The inset table gives the elemental composition of the as-grown films.

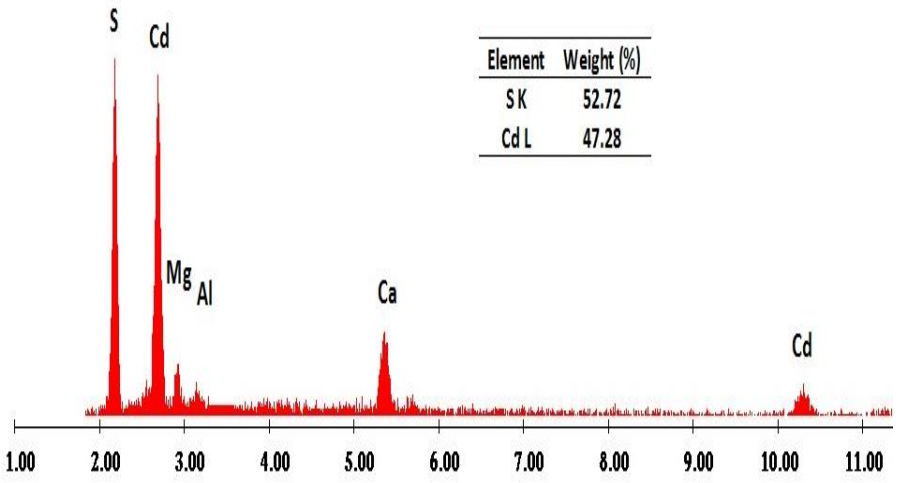
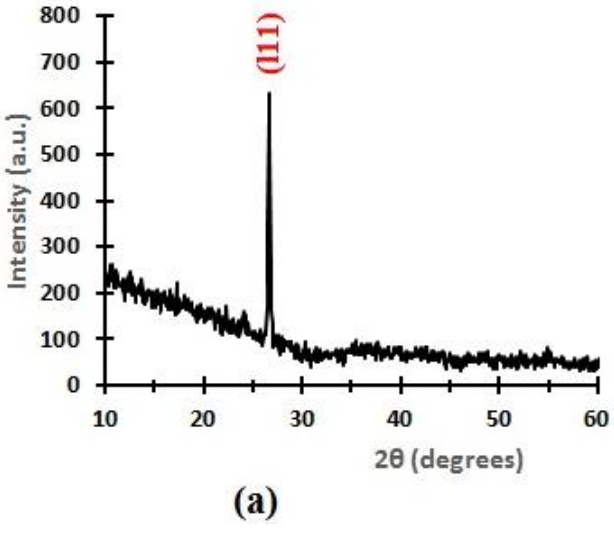


Figure No. 3: EDX of a typical CdS:K film on glass prepared at. 1 % K

Figure 4 presents structural peaks diffracted by the crystal of the samples deposited at different potassium chloride content (a) 1 at.%, (b) 2 at.% and (c) 3 at.%. The diffraction peaks occur at 2θ values about 26.6°, 43.6° and 52.3°corresponding respectively to the (l11), (220), and (311) reflections. The broadening in the XRD peaks realizes that nanoscale CdS:K materials were grown on glass substrate. The position of peaks along with planes show face centered cubic phase as compared with standard reference data (JCPDS Powder Diffraction File (PDF No- 89-0440) [15]. No impurity peaks are detected, revealing the high purity of the as-synthesized samples. The intensity of the predominant peak (111) decreases with increasing K doping content. All the peaks position shifted to lower angles for the doped CdS samples as compared to pure CdS nanoparticles, indicating contraction of the CdS lattice due to the substitutions of K^+ ions into the Cd^+ ions.



(a)

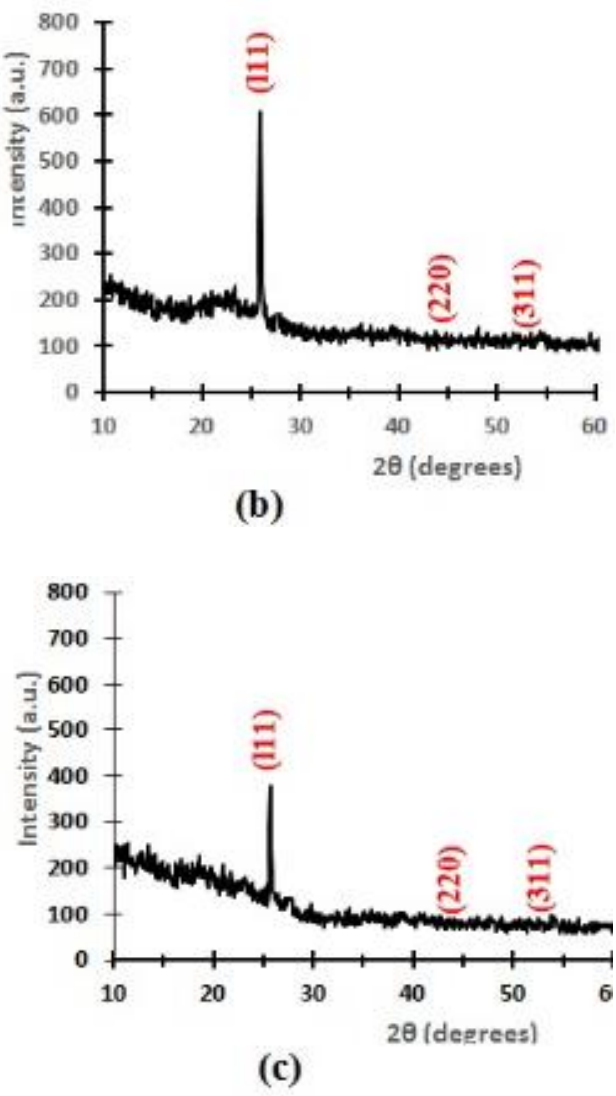


Figure No. 4: XRD patterns of films deposited at different potassium chloride content (a) 1 at.%, (b) 2 at.% and (c) 3 at.%

The intense peak (111) plane was used to calculate the average size of the crystalline domains of crystal grains within a polycrystalline thin film using equation (1). The values obtained were found to be 3.61 nm, 4.11 nm and 4.96 nm respectively. These values agree with the reported results [16] within experimental error.

The optical band gap E_g is calculated from optical measurements by fitting the absorption data to the Tauc relation. We plot the graph $(\alpha h\upsilon)^2 = h\upsilon$ for the films shown in Figure 5. Simply, extrapolation of the linear part of graph $(\alpha h\upsilon)^2 = h\upsilon$ on energy axis at $\alpha=0$ permitted determining the value of the optical band gap E_g (see Figure 4). According to the Tauc relation, the optical band gap E_g is found to be 2.22 eV with increasing changing the

annealing potassium chloride concentration. This change in the optical band gap upon the deposition condition may be explained on the basis of the change in particle size. In fact, the less particle size is associated to the highest the optical energy gap [17].

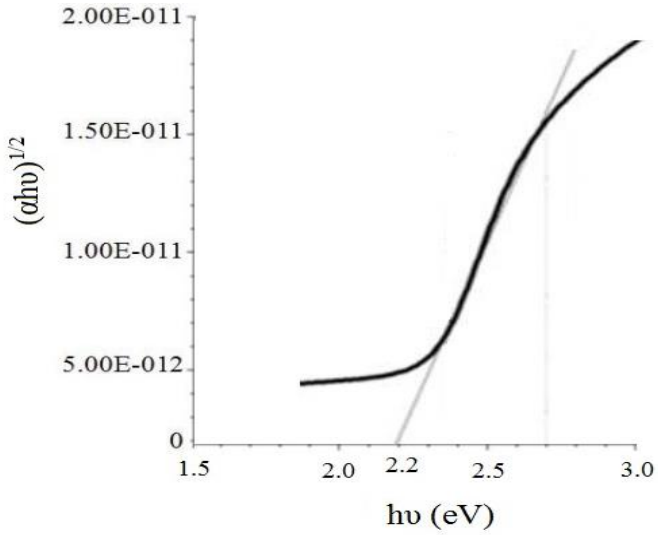


Figure 5: Plot of $(\alpha h\upsilon)^2$ vs. hυ of the CdS:K thin films

CONCLUSION

In summary, potassium chloride cadmium sulphide (CdS:K) thin films with a preferential growth texture (111) have been successfully synthesized by spray pyrolysis technique. Average crystallite size is in the range of 3.61-4.96 nm. Films with homogenous and uniform surface were obtained and composed of Cd and S atomic elements. In addition, films have good adhesion to the glass substrate surface. The optical band gap energy for the as-grown layers was found to be from 2.22 eV to 2.28 eV.

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