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Original research article

Synthesis and characterization of lead sulfide thin films by coumarin assisted CBD method

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ABSTRACT

Coumarin which is a natural substance found in many plants is an organic chemical compound. In this study, we investigated the effects of coumarin adding during the growth of lead sulfide (PbS) thin films by chemical bath deposition (CBD). It was found that significant improvements were observed in the optical and structural properties of the films coated in the presence of coumarin. X-Ray diffraction, UV–vis spectrophotometry and scanning electron microscopy were used to systematically investigate the effect of coumarin on the characteristic properties of the PbS films. X-ray diffraction measurements have revealed that the increasing of coumarin content caused the decreasing the crystallite size of the films. On the other hand, UV–vis analysis showed that the band gap value of the coated samples increased from 1.80 eV to 2.53 eV by changing additive content from 0 to 4%. Also, UV–vis spectra exhibited that the transmittance of the samples varied between 37 and 50%. Further, surface roughness measurements showed that film morphology was influenced by the coumarin content. The results show that coumarin assisted growth of PbS nanostructures is a promising way to improve its physical properties.

1. Introduction

In recent years, metal chalcogenide films have been widely investigated due to their use as semiconducting materials for various applications [1]. Metal chalcogenide materials can be coated on large area and variety of surfaces [2]. Among the various metal chalcogenides, PbS (lead sulfide) is an important IV–VI binary semiconductor with wide range band gap variation (0.41–2.3 eV) [3,4]. Also, properties of lead sulphide like relatively small band gap, abundance in nature, relatively large exciton Bohr radius of 18 nm and high chemical stability makes it desirable and attractive for various applications such as photovoltaic applications [5], gas sensors [6] and infrared detection [7].

Up to now, researches have been used various coating methods to fabricate lead sulfide films including CBD [1], successive ionic layer adsorption and reaction (SILAR) [4], electrodeposition [8] and sol-gel etc. [9]. Among all of these methods, CBD presents various advantages such as simplicity, low temperature, low cost and suitable for large area deposition. To use of PbS thin films in photovoltaic applications, physical properties of the thin films such as surface roughness, crystal structure and particle size are important. It is recorded in the literature that the film properties mentioned above can be tuned by using additive materials in solution growth techniques [10,11].

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In the general sense, it is known that additive materials are added in reaction medium to improve the general quality or to counteract undesirable properties. Coumarin which is an organic chemical compound is one of the additives mentioned in the literature. It is known that coumarin is a natural substance found in many plants. Coumarin has been used as an organic additive in the reaction medium to enhance the nanostructured material quality [10–12]. Hence, due to the potential to improve the physical properties of PbS thin films of the coumarin, we have decide to use the coumarin as an additive material in the growth process. So far, studies on the surface morphology, surface roughness, structural and optical properties of the PbS nanostructures obtained from the bath containing coumarin as additive agent are not reported. In this paper, firstly, we studied the effect of coumarin on the crystal structure, particle size, optical properties and surface roughness of the PbS thin films.

2. Experimental procedure

2.1. Substrate cleaning

In this work, fabrication of PbS thin films was carried out using the CBD method on commercial quality glass substrates (75 mm × 25 mm × 1 mm). Substrates were cleaned with distilled water, acetone and dilute HNO_3 solution (20% v/v) for 10 min respectively. Subsequently, the glass substrates which were washed in deionized water were dried with a hair drier prior to deposition process.

2.2. Sample preparation

All the reagents used in the deposition process were analytical grade (were purchased from Merck). Fabrication of the films (pure or coumarin added) was done in a deposition bath prepared in a 50 mL beaker. The chemical bath contained 0.5 M Lead (II) acetate trihydrate ($Pb(CH_3COO)_2{\cdot}3H_2O$), 1M thiourea ($NH_2CSNH_2$), 1 M triethanolamine ($C_6H_{15}NO_3$) and 1M tri-sodium citrate ($C_6H_5Na_3O_7$). The pH of the chemical solution was adjusted to 12.5 by adding 2 M NaOH. To prepare baths containing coumarin additive, the main solution was poured into five beakers and then 0, 1, 2, 3 and 4 at.% of coumarin were added into each solution. The deposition process was carried out at room temperature and the substrates were taken out from the solution after 18 h as reported elsewhere [1]. After completion of the coating process, the colour of the obtained lead sulphide thin films was dark gray and their image was like mirror.

2.3. Sample characterization

Structural characterization of fabricated samples was made using an X-ray diffractometer (Rigaku Smart Lab XRD). Morphological properties of the pure and coumarin-added films were examined with EVO40-LEO model SEM. Measurements of surface roughness (Ra value) of the samples was made by using a surface roughness tester (Taylor Hobson Surtronic 25). Optical band gap and optical transmittance were calculated using the data obtained from UV–vis spectrophotometer (Thermo Scientific Evolution 160).

3. Results and discussion

3.1. Structural properties

The crystallinity and preferred orientations of the samples were studied by analysis of the XRD patterns. XRD patterns of the cubic PbS nanostructures obtained from the bath containing coumarin as additive agent are shown in Fig. 1. The pattern of PbS thin film without coumarin was also included in the figure. The reflection peaks at $2\theta \approx 25.93^\circ$, 30.00°, 42.95°, 50.90°, 53.33°, 62.43°, 68.85°, 70.83°, 78.80° and 84.72° corresponding to (111), (200), (220), (311), (222), (400), (331), (420), (422) and (511) orientation of PbS has been observed in all the chalcogenide samples. It can be seen from the figure that all diffraction peaks are in good agreement with standard data of Joint Committee on Powder Diffraction Standards (JCPDS: 05-0592) data. Also, chalcogenide nanostructures are polycrystalline state and the results indicated that the orientation of crystals is mainly in (200) and (111) direction. Other observed peaks are less intense as compared to (200) and (111). Furthermore, any peak referring to impurity phases were not detected in the patterns. On the other hand, similar studies in the literature have not been reported any coumarin peak in XRD analysis [10,16]. The reason for this may be that coumarin can effect only on the nucleation mechanism during film formation process without introduce into lattice. From the XRD results it was clearly seen that coumarin concentration in the growth solution has a small decreasing effect on the intensity of the peaks especially corresponding to (200) and (111) planes. This behaviour can be attributed to additive induced some structural disorder in the films [2]. The recorded (200) and (111) peak intensities of the films are listed in Table 1.

It is known that texture is the distribution of crystallographic orientations of a polycrystalline sample. Table 1 displays the values of the texture coefficient $TC_{(hkl)}$ for four different planes. The addition of additive agent in the growth solution may be induces the change of diffusion rate of cationic and anionic precursors at the surface during growth process and this