Effect of Aqueous Solution Molarity on the Structural and Electrical Properties of Spray Pyrolysed Lead Sulfide (PbS) Thin Films

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ABSTRACT. Lead sulfide (PbS) thin films of different molarities (0.05 M, 0.075 M and 0.1 M) were prepared on glass substrates at 325 °C by chemical spray pyrolysis (CSP) technique. X-ray diffraction patterns confirm the proper phase formation of the PbS. The X-ray diffraction patterns' results reveal that the all of PbS films have a face centered cubic structure with preferential reflection of (200) plane. The crystallite grain size was calculated using Scherrer formula and it is found that the 0.1M has maximum crystallite grain size (37.4 nm). Depending on the molarity, Hall measurement showed that the electrical resistivity and mobility at room temperature varied in the range $6.3 \times 10^3 \Omega \cdot \text{cm}$ to $2.1 \times 10^3 \Omega \cdot \text{cm}$ and $4.79 \text{ cm}^2/\text{V.S}$ to $24.3 \text{ cm}^2/\text{V.S}$.

1. INTRODUCTION

Lead sulfide (PbS) is a compound semiconductor of IV-VI group which has a lattice with unit cell face center cube [1]. PbS is an important direct narrow gap semiconductor material with an approximate energy band gap energy of 0.4 eV at 300 K and a relatively large excitation Bohr radius of 18 nm [2,3]. These two properties make PbS very suitable for in farad detection and solar cell applications [4].

A number of techniques have been used by various authors to deposit PbS thin films including evaporation [5], photoaccelerated chemical deposition [6], chemical bath deposition [7] and chemical spray pyrolysis [8]. Among these different techniques, spray technique is advantageous on account of the low cost and its suitability for forming large area thin films. In this present work, an attempt has been made to study the structural and electrical properties of the PbS thin films with different molarities (0.05 M, 0.075 M and 0.1 M) by Chemical Spray Pyrolysis technique on glass substrate.

2. EXPERIMENTAL PROCEDURE

2.1. Substrates cleaning

Glass slides with the dimensions of $75 \times 25 \times 2$ mm were used as substrates. Firstly, the substrates were cleaned by alcohol for 10 min to remove contamination. After the cleaning process, all of the substrates were rinsed with distilled water. The samples were dried with nitrogen (N_2) gas.

2.2. Materials

Lead (II) acetate trihydrate ($\text{Pb}(\text{CH}_3\text{CO}_2)_2 \cdot 3\text{H}_2\text{O}$; 99.999%), Thiourea ($\text{CS}(\text{NH}_2)_2$, $\geq 99.0\%$) were acquired from Acros Organics.

2.3. Preparation of PbS Solutions

In the preparation of PbS films, aqueous solutions of Lead (II) acetate trihydrate ($\text{Pb}(\text{CH}_3\text{CO}_2)_2 \cdot 3\text{H}_2\text{O}$) and Thiourea ($\text{CS}(\text{NH}_2)_2$) with different molarities (0.05M, 0.075 M and 0.1 M) and appropriate volumes were mixed with distilled water by using magnetic stirrer for 30 minutes and the resultant solution was sprayed on glass substrates which were kept at temperature of 325 °C. In order to get uniform thin films, the height of the spraying nozzle and the rate of spray process were

kept constant during the deposition process at 35 cm and 15 cm³/min. The spraying process lasted about 6 second. The period between spraying processes was about 1 min; this period was enough to avoid excessive cooling of the substrate. The thickness of the prepared films was about 400 nm measured by gravimetric method. The crystallographic structure of the PbS thin films deposited on the glass substrates was determined with a high resolution X-ray diffractometer system (Model: Panalytical Empyrean) with CuK α radiation (λ) of 0.154 nm. The electrical properties of PbS thin films was measured by Hall measurements (HL5500PC) system.

3. RESULTS AND DISCUSSION

3.1 STRUCTURAL PROPERTIES

Fig. 1 shows the X-ray diffraction pattern of as deposited PbS with different concentration are shown in **Fig.1**. All PbS films have cubic rock salt (NaCl) type structures. XRD patterns of all the PbS thin films showed sharp (1 1 1) and (2 0 0) peaks along with minor peaks of (2 2 0), (3 1 1) and (2 2 2) planes to cubic structure of PbS thin films, as confirmed by standard ASTM card (No. 030660020). The strongest peak occurs at $2\theta \sim 30^\circ$ which is referred to (200) plane. The molarity had a significant effect on the X-ray diffraction structural analysis of PbS thin films as shown in **Fig.1**. As the concentration of the precursor increases there is an increase in intensity of (200) plane. These results are in accordance with previously reported [9]. It can be noticed that 2θ for (200) direction decrease with molarity increasing, as shown in **Table (1)**. The crystalline grain size (t) of the PbS films was determined with the Scherrer formula [10].

$$t = \frac{0.9\lambda}{\beta \cos\theta} \quad (3.1)$$

where β is the full width at half maximum (FWHM) of the peak, λ is the wavelength of the X-ray, 1.5406 Å, and θ is the peak position. Based upon the line width of the (200) diffraction peak, the values of crystalline grain size are shown in **Table (1)**. That is, the grain size of the PbS films increased with increasing molarity. This small grain size of the film is reveals that the nanocrystalline nature of the film. For the films grown from concentrations 0.05 M to 0.1 M, the peak position shifted to lower angles indicating increase of Inter Planar distance (d) value and hence the bond length [11].

Fig. 1. XRD pattern of PbS thin films at different molarities.

Table 1. Structural parameters of PbS thin films at different molarities.

Molarity (M)	0.05	0.075	0.1
2θ (deg)	30.0738	30.0481	30.0351
<i>hkl</i>	(200)	(200)	(200)
Inter Planar distance (<i>d</i>) (Å)	2.97153	2.97401	2.97528
Crystalline grain size (<i>t</i>) (nm)	26.3	28.4	37.4

3.2 ELECTRICAL PROPERTIES

Table 2 shows the dependence of the electrical resistivity and Hall mobility on the solution concentration. The decrease in the resistivity of the PbS thin film with concentration of precursor **0.1M** compared to that of the PbS thin film with concentration of precursor **0.05M** due to a increase in the Hall mobility. This behavior agrees with previous reports [12].

Table 2. Measured electrical parameters for PbS thin films at different molarities.

<i>Sample</i>	<i>Resistivity (ρ) ($\Omega.cm$)</i>	<i>Hall mobility (μ_H) ($cm^2/V.S$)</i>
0.05M	6.3×10^3	4.79
0.075M	4.7×10^3	12.23
0.1M	2.1×10^3	24.3

4. CONCLUSIONS

In summary, PbS thin films were prepared successfully by chemical spray pyrolysis technique on glass substrates at 325°C. Effects of reactant concentration on structural and electrical properties of the PbS films were studied. The XRD results showed that all films have cubic rock salt (NaCl) type structures. PbS films deposited with higher concentration of precursor solution shows clear improvement in crystallinity. The crystallite grain size of PbS thin films increases with molarity increasing. From the electrical properties, it was found that the Hall mobility increases as the molarity increases and the Hall mobility values range between 4.79 and 24.3 ($cm^2/V.S$).

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