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Studies on growth and characterization of ternary $CdS_{1-x}Se_x$ alloy thin films deposited by chemical bath deposition technique

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ABSTRACT

Ternary alloyed $CdS_{1-x}Se_x$ thin films of variable composition '*x*' were grown by the simple and economical chemical bath deposition technique. The as-grown thin films were characterized for structural, compositional, surface morphological, optical and electrical studies. The X-ray diffraction (XRD) patterns of the sample indicated that all the samples were polycrystalline in nature with hexagonal structure. Scanning electron microscopy (SEM) micrographs showed uniform morphology with spherical shaped grains distributed over entire glass substrate. EDAX studies confirmed that the $CdS_{1-x}Se_x$ films were having approximately same stoichiometry initially as well as finally. Room temperature optical measurements showed that band gap engineering could be realized in $CdS_{1-x}Se_x$ thin films via modulation in composition '*x*'. Electrical resistivity of $CdS_{1-x}Se_x$ thin films for various compositions was found to be low. The broad and fine tunable band gap properties of ternary $CdS_{1-x}Se_x$ thin films have potential applications in opto-electronic devices.

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1. Introduction

Group II–VI semiconductors with energy gaps covering the visible spectral range are promising candidates for opto-electronic devices [1,2]. CdS and CdSe are two very important wide gap semiconductors, because of their wide applications in opto-electronics, such as non-linear optics, visible-light emitting diodes and lasers [3]. However, for some opto-electronic applications it is important to be able to tune the emission wavelength. The tunability can be achieved through composition modulation, for example, alloyed II–VI semiconductor ternary of $CdS_{1-x}Se_x$, $Zn_xMg_{1-x}O$, and $Cd_{1-x}Mn_xS$ with continuously tuned band gap have been reported [4–8]. Consequently, wavelength tunable emission can be achieved from ternary compounds by simple adjustment of composition. The alloy of CdSSe should have more applications since its band gap can be tuned by means of the composition in between ∼2.44 eV (for CdS) and ∼1.72 eV (for CdSe), almost covering the entire visible range. However, for the efficient opto-electronic device application, material is usually between CdS in which very high sensitivity is possible but response time is high and CdSSe in which a lower response time is possible at the cost of some loss in sensitivity [9].

Considering the advantages laid down by the ternary alloy $CdS_{1-x}Se_x$ thin films, we focus our work to investigate the tunable band gap properties of this alloy. For this we synthesize the ternary $CdS_{1-x}Se_x$ alloy in thin film form using chemical bath deposition (CBD) technique with variable composition '*x*' ($0.0 \leq x \leq 1.0$). The as-grown films were studied for structural, compositional, morphological, optical and electrical properties and the results obtained are compared with literature wherever necessary.

2. Experimental details

2.1. Thin film preparation

For the preparation of $CdS_{1-x}Se_x$ films, 0.1 M solution of cadmium chloride, thiourea and sodium selenosulphite were prepared in double distilled water. A solution of sodium

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selenosulphite was prepared by refluxing 100 ml of 0.1 M sodium sulphite with selenium powder for about 5–6 h. This solution will contain excess of sodium sulphite which prevents the oxidation of selenide to selenium [10].

2.1.1. For the deposition of CdS

Equal volume (20 ml), molarity (0.1 M) of cadmium chloride and thiourea were taken. The pH of the resultant mixture was kept ~11.5 by addition of aqueous ammonia with the constant rate (2 ml/min) and constant stirring. The temperature of the reaction bath was maintained at 80 ± 5 °C. The deposition was carried out for a period of 2 h.

2.1.2. For the deposition of CdSe

Similar procedure was employed to deposit CdSe. However, instead of thiourea, sodium selenosulphite was used. The deposition was carried for a period of 1 h.

2.1.3. For the deposition of $CdS_{1-x}Se_x$

Similar procedure was employed to deposit $CdS_{1-x}Se_x$ thin films. However, here the volume of cadmium chloride was kept fixed (20 ml), while the volumes of thiourea and sodium selenosulphite were varied. To maintain the pH of the solution aqueous ammonia solution was added proportionally in the above solution bath. The temperature of the reaction bath was maintained at 80 ± 5 °C. For achieving same thickness (0.32 μm) the time duration of deposition for various compositions was different. Table 1 summarizes the various parameters for the deposition of $CdS_{1-x}Se_x$ thin films.

2.2. Characterization technique

The ternary alloy $CdS_{1-x}Se_x$ thin films were characterized for structural, compositional, morphological, optical and electrical properties. The film thickness was measured using Fizeau Fringe technique. X-ray diffraction (XRD) patterns of the films were recorded on Brucker AXS, Germany (D8 Advanced) diffractometer in the scanning range 20–70° (2θ) using Cu Kα radiations with wavelength 1.5405 Å. The surface morphology and compositional analysis was done using JOEL–JSM 5600. To study the optical characteristic of the film absorbance spectra were recorded in the range 350–850 nm by means of PerkinElmer UV–vis spectrophotometer Lambda 25. The room temperature resistivity was measured using *I–V* characteristics (Lab equipment unit {model no. 2004} interfaced with computer), over the range from ±3 V. Silver paint was employed to ensure good ohmic contacts with the film.

3. Results and discussion

3.1. Growth mechanism

The solution chemistry involved in the deposition of $CdS_{1-x}Se_x$ thin films takes place by the formation of chalcogen ions by the process of hydrolysis in alkaline medium. As a result, the metal ions get combined with the chalcogen ions forming the desired compound.

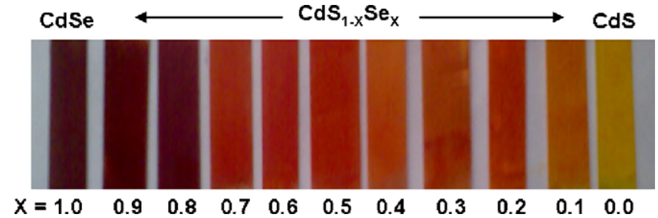

Fig. 1. Thin film images for $CdS_{1-x}Se_x$, of different compositions '*x*' ($0.0 \le x \le 1.0$).

The deposition process is based on the slow release of Cd^{2+}, S^{2-} and Se^{2-} ions. The amount of sulphite and selenide ions in the bath is controlled through setting up of appropriate chemical equilibrium [11,12], i.e., by adjusting properly the bath parameters and considering the solubility product, etc. (as discussed in Section 2.1). For the deposition of film, the availability of the nucleation center over the substrate is necessary. Such centers are normally formed through the adsorption of metal hydroxo species over the surface. The hydroxo group would be substituted by the sulphite and selenide ions, which would thereby, form an initial layer of the metal chalcogenide [13]. The initial layer acts as catalytic surface and this surface is crucial for the uniform deposition of thin film. On this surface, the deposition of the thin film takes place through the condensation of the metal sulphite and selenide ions, which acts as a catalytic surface. The formation of a film can take place heterogeneously on the substrate surface or homogeneously in the solution producing precipitate. The homogeneous process is highly undesirable since the adsorption of the particles on the substrate surface yields powdery and non-adherent films. Hence the homogeneous process can be suppressed by using high rate of formation, i.e., by setting low concentrations of reactants, high concentrations of aqueous ammonia, etc. In our case, a predominant heterogeneous reaction was characterized by a clear reaction bath and the films formed were well adherent and without powdery. The visual observation of the films as we go from CdS to CdSe is shown in Fig. 1. It can be clearly observed that the yellowish color CdS thin film changes to dark brownish CdSe thin films with the change in the composition '*x*' ($0.0 \le x \le 1.0$).

A simple reaction mechanism can be written as follows:

The sulphite ions are released due to decomposition of thiourea in an alkaline medium as,

$$SC(NH_2)_2 + 3OH^- \rightarrow 2NH_3 + CO_3{}^{2-} + HS^- \quad (1)$$

$$HS^- + OH^- \rightarrow S^{2-} + H_2O \quad (2)$$

Similarly, the selenide ions are released as,

$$Na_2SeSO_3 + OH^- \rightarrow Na_2SO_4 + HSe^- \quad (3)$$

$$HSe^- + OH^- \rightarrow Se^{2-} + H_2O \quad (4)$$

Table 1
Compositional quantities for $CdS_{1-x}Se_x$ films initially taken in the solution bath

Composition '*x*'	Solution of $CdCl_2$ (ml)	Solution of $(CS)[NH_2]_2$ (ml)	Solution of Na_2SeSO_3 (ml)	pH of bath	Deposition time (min)
0.0	20	20	00	11.5	120
0.2	20	16	04	11.2	108
0.4	20	12	08	10.9	96
0.6	20	08	12	10.6	84
0.8	20	04	16	10.3	72
1.0	20	00	20	10.0	60

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