Enhancement the sensitivity of CdS nano structure by adding of rare earth materials

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Abstract. Thin films of Cerium (Ce³⁺) doped Cadmium Sulfide (CdS) were successfully deposited on glass and silicon substrates by Chemical Spray Pyrolysis Method at different ratios with temperature 200 oC. The results shown a high quality nanoparticles and a strong orientation to the (002) plane of the wurtzite (hexagonal) phase. The fast rise and decay times of the photocurrent were confirmed that the best quality of the fabricate photodetector device. Light responsivity was at (460, 550 and 680 nm), and the device showed a good specific detectivity and quantum efficiency when illuminated by (460, 550 and 680 nm). CdS:Ce at 10% clarity higher responsivity (0.144 A/W), quantum efficiency (38.58%), and specific detectivity (2.7×10¹¹ Jones) at 460 nm illumination (3.108 mW/cm²), also the device for CdS:Ce at 10% reveals the fast with 800 ms rise time and 860 ms decay time and the sensitivity reach to 5523.

1. Introduction

Cadmium sulfide (CdS) is attracts the attention of several research groups [1]. It has a direct energy gap of 2.42 eV at room temperature [2] with a low work function of approximately 4.2 eV [3]. It was found that the doping process and the type of dopants play crucial roles in the enhancements of the electrical and optical properties of the CdS. Several dopants specifically the rare earth metals were studied intensively [4]. The electronic structure of rare earth elements are the same in their outer of (5s² 5p⁶ 6s²) and differ only by electrons occupying the inner partially filled 4f shell [5], and their Ionization occurs when they lose two 6s and one 4f electrons and form a stable trivalent state [6]. Doping of rare earth element like cerium clearly affects the particle size where it is reduce and also increases surface area [7]. One of the most common element among rare elements is cerium, which has strong acidity, strong oxidation, high mechanical and optical properties and it's retractable [8]. Recently, large numbers of CdS nanostructures [9-14], have been used to fabricate photodetectors by using different methods except chemical spray pyrolysis method. In this work we report the effect of Ce doping on the photodetector (in UV - Visible range), performance based on CdS will be explore, and synthesized by chemical Spray Pyrolysis Method, which characterized by no vacuum system need for deposition, consider inexpensive and simplicity method, quite the opposite of the complexity of other methods of preparation [15,16,17].

2. EXPERIMENTAL WORK

To prepare chemical solution for pure and doped CdS, 0.1 M from cadmium chloride (CdCl₂·H₂O) and 0.2 M from thiourea (NH₂SCNH₂) as a source of sulphur ion were separately dissolved in 50 ml of distilled water and 10 ml ethanol. The material supply from THOMAS BAKER company with purity 99.9%. Cadmium chloride solution is mixed with thiourea using a magnetic stirrer at room temperature for 30 minutes to complete the solubility process. For doping process Cerium Oxide (CeO₂) is dissolved at doping ratios of (10, 20 and 30) wt.% with cadmium source using a magnetic stirrer for 30 minutes and the
mixture is added to sulphur ion solution to get CdS:Ce solution. This final solution is transferred to spray container. To thin films preparation, chemical spray pyrolysis method (CSP) was used to prepare the thin films with parameters: substrate temperature at 200°C, the distance between the nozzle and substrates was set at 20 cm, flow rate was 0.6 ml/min. The spray process continued until getting the required thickness of 200 nm.

For electrical contact, aluminium deposited on the surface of CdS pure and CdS:Ce thin films by thermal evaporation technique under vacuum (10-5 mbar), using Edward coating unit model (306 A).

The prepared thin films were characterized for their phase identification using X-Ray diffraction unit model MiniFlex II (from Rigaku) and the surface morphology was characterized via field emission scanning electron microscope (FESEM) (from Field Electron and Ion, FEI) (Nova nanoSEM 450) the Netherlands. The optical absorbance spectra were measured using UV/Visible SP – 8001 spectrophotometer over the range 190–1100 nm. The output data of wavelength and absorbance are used in a computer program to deduce the optical energy band gap and transmission. The photoluminescence and the photoconductivity investigated by (RF-551) spectrofluorometric detector (Shimadzu, Japan) and the measurements are done by using sensitive digital electrometer type Keithley (2400).

3. RESULTS AND DISCUSSION

Thin films structural properties were examined by the XRD as shown in Fig. (1). CdS sample has polycrystalline structure with the reflection planes of (100), (002), (101), (110), (103), and (112), all those reflection planes noted also for CdS:Ce, and all patterns show the film exhibited hexagonal (wurtzite) crystal structure as indicated by the absence of characteristic (200) and (311) peaks of the cubic CdS structure, with a preferential orientation along the (002) plane. From the XRD spectra it was also observed that no diffraction peaks corresponding to the impurity phases were detected and this rule out Ce deposition or secondary phases, which similar to Sreenivas et al.[7] and that confirmed the successful incorporation of Ce ions into the crystal lattice of CdS particles[18]. The average crystallite size calculated by Debye Scherrer’s equation [19]:

\[ D = \frac{0.89 \lambda}{\beta \cos \theta} \]  

where \( \lambda \) is a wavelength of the X rays, \( \beta \) is the full width at half and \( \theta \) is the diffraction angle.

The averages crystallite size were found to be decrease with increase of doping and the data of pure and doped CdS listed in Table (1).

Thin films morphological properties were tested by the FESEM images. Fig.(2)(a-d) show the images of FESEM for CdS and doped CdS:Ce at 10, 20 and 30%. Image (a) thin film, grains are somewhat homogeneous in both shape and size and grain boundaries are very clear because of the big grain size growth is due to the agglomeration of individual particles and the surface covered by big grains.

For minimum and maximum (10 and 30%) Ce doped CdS as show in images (b,d), it is observed the topography is different than that of pure CdS, with a compact and dense structure and better in grain correlation, the incorporation of cerium caused the grain size to decrease with almost uniform distribution over the surface, and the grain boundaries to disappear significantly, which refer to better for CdS thin films for photovoltaic applications [20] and this agreement with hurma [21].

While in image (c) has different topography content grains like rods ordered randomly and the grain size bigger with vacancies appeared clearly.

UV-visible transmission spectrum for all simples are revealed in Fig.(3). It is observed that the transmission for all simples are high in visible and near IR region, the value of the transmission was (95%, 90%, 84% and 60%) CdS:Ce at (30, 10 and 20%) and pure CdS respectively, that mean highly transparent is found for doped thin film, it can be used in solar cell as transparent window.
The optical band gap of the doped and undoped CdS thin films, as shown in Fig. (4) was estimated by Tauc plot. For pure CdS optical band gap equal to (2.57 eV), while CdS:Ce at (10, 20 and 30%) equal to (2.65, 2.62 and 2.66 eV), this slightly increase in the energy gap is refers to the nano phase for undoped and doped CdS. The Increase band gap value may be due to increasing in quantum effect which it result of decreasing in the particle size [22].

Figure (5) shows the PL results of CdS and CdS:Ce at excitation wavelength 400 nm. For pure CdS there is a broad defect emission because the present of point defects such as cadmium vacancies (V_{Cd}), sulfur vacancies (V_{S}), cadmium interstitials (I_{Cd}) and sulphur interstitials (I_{S}), which act as luminescent centers [23]. It is observed that three peaks, a broad peak between (320 and 480) nm, which centered at 440 nm. The strong PL emission indicates the high crystallinity of CdS sample, and consistent with the XRD results[24], the second peak centered at 530, and the third peak centered at 610nm, with UV, green and orange emission respectively. The UV emission for the first peak as shown in figure, attributed to transitions from the deep and shallow states [25]. The green emission band of the second peak is due to electronic transition from conduction band to an acceptor level due to interstitial sulphur ions (I_{S}) [26]. The origin of orange band is originating from the transition from the donor levels, created by the occupation interstitial sites of Cd atoms (I_{Cd}), to the valence band [27]. The peaks position of CdS:Ce emission slightly shifts toward longer wavelength region with higher intensity are associated with increase of doping concentrations, Thus it confirms that luminescence property of CdS nanoparticles enhanced when Ce+3 was introduced into the CdS. Maleki et al. [28].

The characteristic of time I–t curves for the undoped and doped CdS photodetector as shown in Fig. (6) was studied by periodic exposed under the selected wavelengths of (460, 550 and 680nm) with an illumination intensities (3.108, 2.970 and 3.044 mW/cm²) respectively under bias voltage (0 V). In the dark the current value decreased and jumped to a steady higher value under illumination, CdS:Ce at 10% has the highest value of (I_{on}/I_{off}) ratios (56.18, 38.46 and 10) at wavelengths (460, 550 and 680 nm) respectively. The average rise and decay times for all samples are (0.8000 and 0.8060s) respectively and this fast response is reported by Zhao et al. [29], and Liang Guo et al. [30]. The short decay time refers to the high surface to volume ratio, which caused dangling bonds and defects and results of that increase in recombine between electron and hole pairs in dark [24]. The sensitivity of the fabricated device calculated by the following relationship [31]:

\[ S = \frac{I_{light} - I_{dark}}{I_{dark}} \times 100\% \quad (2) \]

where \(I_{light}\) and \(I_{dark}\) represent the photocurrents under illumination and in the dark respectively. In our work the sensitivity was higher than reported by Liang Guo et al. [30] and Mahdi et al.[24], and proved photodetector can generate photocurrent by light exposure without applying any bias voltage as solar cell.

Fig. (7) shows the responsivity (R_{λ}) of the photodetectors which calculated by equation[29]:

\[ R_{λ} = \frac{I_{light} - I_{dark}}{P_{exc}} \quad (3) \]

\[ P_{exc} = I_{light} \cdot A \quad (4) \]

Where \(P_{exc}, I_{light}\) and \(A\) are the excitation power, illumination density, and the illuminated device area respectively.

It can be observed the maximum value of the spectral responsivity is for device content 10%Ce at 460 nm. Also we noted that the spectral responsivity began decreasing with the increase of doping concentration and wavelength. Because of the band gap structure for undoped and doped CdS (2.57 – 2.66 eV) we notes that the responsivity indicated that the 460 nm have a better response wavelength than 550 and 680 nm, revealing that the response
spectrum is directly related to the band gap values [32]. The responsivity value of CdS:Ce at 10% is high than that reported in ref.[33].

Figure (8) shows the quantum efficiency (QE) of undoped and doped CdS with Ce different ratios, which is determined as a function of wavelength by using the following relationship [29]:

\[ \text{QE} = \frac{h \cdot c \cdot R_a}{e \cdot \lambda} \quad \text{(5)} \]

The photodetector CdS:Ce at 10% has maximum value of the (QE), it's equal to 38.58% at 460 nm wavelength, this value is high comparable to that reported in ref.[34].

The specific detectivity (D*) for pure CdS and CdS:Ce at different concentration ratios, using the following equation to calculate the (D*)[29]:

\[ D^* = \frac{R_s \sqrt{R}}{2 \pi I_{dark}} \quad \text{(6)} \]

The maximum value of (D*) is for CdS:Ce at 10%, is equal to \(2.83 \times 10^{11}\) Jones as shown in Fig. (9). Specific detectivity of our device is much higher compared to the detectivity which reported photodetector based on inorganic ZnS hybrid system \(1 \times 10^{-10}\) Jones [35]. All data of sensitivity (S), responsivity (R), quantum efficiency (QE) and specific detectivity (D*) for CdS:Ce photodetector were listed in Table (2).

4. Conclusions

CdS and CdS:Ce at different Ce ratios prepared by chemical spray pyrolysis method are used as photodetectors. X-ray diffraction measurements show polycrystalline with hexagonal structural, the FESEM images point to the surface is covered by big grains with a triangular shape, and the optical measurements explain enhancement in transmission and photoluminescence for doped CdS compared with pure CdS, and all samples have direct energy gap. The photoconductance of the photodetectors were studied, which showed higher response at 460, 550, and 680 nm. The device for CdS:Ce at 10% clarifies higher responsivity, quantum efficiency and specific detectivity at 460 nm with fast response and decay time compared with the other Ce concentrations. The stability of these devices for a long period of time could be good candidates for high performance photodetectors in various applications.

Reference

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Table 1: Structural parameters for pure CdS and CdS:Ce at different ratio

| Sample | 2θ (Deg.) | FWHM (Deg.) | dhkl | Exp.(Å) | G.S (nm) | dhkl Std.(Å) | Phase       | hkl  |
|--------|-----------|-------------|------|---------|----------|--------------|-------------|------|
|        | 25.0525   | 0.1690      | 3.5516 | 48.2    | 3.5940   | Hex. CdS    | (100)      |      |
| Pure   | 26.7571   | 0.2010      | 3.3291 | 40.6    | 3.3685   | Hex. CdS    | (002)      |      |
|        | 28.4310   | 0.2457      | 3.1368 | 33.4    | 3.1710   | Hex. CdS    | (101)      |      |
|        | 43.9724   | 0.2457      | 2.0575 | 34.9    | 2.0750   | Hex. CdS    | (110)      |      |
|        | 48.1495   | 0.3013      | 1.8883 | 28.9    | 1.9045   | Hex. CdS    | (103)      |      |
|        | 52.1577   | 0.3072      | 1.7522 | 28.8    | 1.7667   | Hex. CdS    | (112)      |      |
| Ce 10% | 24.9450   | 0.2304      | 3.5667 | 35.3    | 3.5940   | Hex. CdS    | (100)      |      |
|        | 26.6650   | 0.2611      | 3.3404 | 31.3    | 3.3685   | Hex. CdS    | (002)      |      |
|        | 28.3542   | 0.2610      | 3.1451 | 31.4    | 3.1710   | Hex. CdS    | (101)      |      |
|        | 43.8649   | 0.3071      | 2.0623 | 27.9    | 2.0750   | Hex. CdS    | (110)      |      |
|        | 48.0727   | 0.3379      | 1.8912 | 25.7    | 1.9045   | Hex. CdS    | (103)      |      |
|        | 52.0502   | 0.3072      | 1.7556 | 28.8    | 1.7667   | Hex. CdS    | (112)      |      |
| Ce 20% | 25.0316   | 0.2848      | 3.5545 | 28.6    | 3.5940   | Hex. CdS    | (100)      |      |
|        | 26.7880   | 0.2848      | 3.3253 | 28.7    | 3.3685   | Hex. CdS    | (002)      |      |
|        | 28.4494   | 0.2848      | 3.1348 | 28.8    | 3.1710   | Hex. CdS    | (101)      |      |
|        | 43.9715   | 0.2848      | 2.0576 | 30.1    | 2.0750   | Hex. CdS    | (110)      |      |
|        | 48.0538   | 0.3797      | 1.8919 | 22.9    | 1.9045   | Hex. CdS    | (103)      |      |
|        | 52.1361   | 0.2848      | 1.7529 | 31.1    | 1.7667   | Hex. CdS    | (112)      |      |
| Ce 30% | 24.9367   | 0.2848      | 3.5679 | 28.6    | 3.5940   | Hex. CdS    | (100)      |      |
|        | 26.6930   | 0.2848      | 3.3370 | 28.7    | 3.3685   | Hex. CdS    | (002)      |      |
|        | 28.3544   | 0.2848      | 3.1451 | 28.8    | 3.1710   | Hex. CdS    | (101)      |      |
|        | 43.8766   | 0.3323      | 2.0618 | 25.8    | 2.0750   | Hex. CdS    | (110)      |      |
|        | 48.1013   | 0.3798      | 1.8901 | 22.9    | 1.9045   | Hex. CdS    | (103)      |      |
|        | 52.0886   | 0.3797      | 1.7544 | 23.3    | 1.7667   | Hex. CdS    | (112)      |      |

Table (2) S%, Rλ, QE and D* for pure CdS and CdS:Ce at different Ce ratios at selected wavelengths.

| Details | 400 nm | 550 nm | 600 nm |
|---------|--------|--------|--------|
|         | S%     | R(A/W) | QE %   | D* Jones |
|         | 560    | 0.0097 | 0.0092 | 3.3×1010 |
|         | 5523   | 0.1440 | 0.1250 | 2.7×1011 |
|         | 331    | 0.0245 | 0.0240 | 1.26×1011 |
|         | 84     | 0.0153 | 0.0137 | 1.9×1010 |
|         | 45     | 0.0137 | 0.0133 | 1.9×1010 |
|         | 550    | 4.19   | 4.19   | 1.7×1010 |
|         | 45     | 2.37   | 2.37   | 1.7×1010 |

|         | 163    | 0.082  | 2.9×1010 |
|         | 827    | 0.114  | 2.2×1011 |
|         | 521    | 0.0230 | 1.19×1010 |
|         | 28     | 4.19   | 1.7×1010 |
|         | 2.37   | 4.19   | 1.7×1010 |
|         | 4.19   | 4.19   | 1.7×1010 |
Fig. (1): X-Ray diffraction patterns for CdS and CdS:Ce with doping ratio 10, 20 and 30%.

Fig. (2): FESEM image of (a) pure CdS (b) CdS:Ce 10% (c) CdS:Ce 20% (d) CdS:Ce 30%.
Fig. (3) : Transmission for CdS and CdS:Ce thin films at different ratio.

Fig. (4) optical energy gap for CdS and CdS:Ce at different ratio.

Fig. (5) PL spectrum of pure CdS and CdS:Ce at excitation wavelength 400 nm.
Fig. (6) I–t curves for the undoped and doped CdS photodetector at selected wavelengths.
Fig. (7) Responsivity of CdS and CdS:Ce at different Ce ratios.

Fig. (8) Quantum Efficiency of CdS and CdS:Ce at different Ce ratios.

Fig. (9) Specific Detectivity of CdS and CdS:Ce at different Ce ratios.