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Original Research Article

Effect of deposition potential on the structural and optical properties of electrodeposited antimony doped zinc telluride (Sb:ZnTe) thin films for optoelectronic applications

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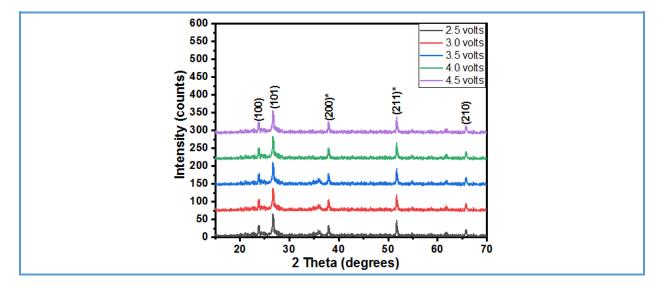
KEYWORDS

Sb:ZnTe thin film Optoelectronic materials Electrodeposition Semiconductor alloys Energy band engineering

ABSTRACT

Thin films of antimony-doped zinc telluride (Sb:ZnTe) were produced using an electrodeposition method. The examination included both the structural and optical characteristics, as well as the size of the crystallites. X-ray diffraction (XRD) analysis revealed Sb:ZnTe on Fluorine doped Tin Oxide (FTO) substrates had a hexagonal structure. The crystallite sizes in these thin films vary between 23.88 nm and 33.00 nm with dislocation density within the range of 2.32 to $1.02 \times 10^{15} lines/m^2$ and the microstrain between 5.59 and 3.99. UV-Vis spectroscopy revealed the absorbance values of the film decreased, with a range of 33% to 54% as the wavelength increased from 400 nm to 1100 nm. The transmittance and reflectance values of the film varied between 28.50% and 48.0% and less than 20.5%, respectively, suggesting that the deposited thin films are appropriate for use as antireflective coatings in smart window technology. The refractive indices of the films varied between 2.40 and 2.63. The extinction coefficient was found to increase with wavelength across the studied spectrum (400 nm to 1100 nm) and decrease with higher deposition potential. These extinction coefficient values suggest that the films are appropriate for use as absorber layers in thin-film solar cells. A band gap of 2.00 eV was determined at 2.5 volts and as the deposition potential increased, the films showed a decrease in the energy band gap. Gravimetric method analysis revealed that the thickness of antimony doped zinc telluride films increased from 126.18 nm to 378 nm as the deposition potentials increased from 2.5 volts to 4.5 volts. It is critical to control the electrodeposition potential to achieve the desired film thickness and properties.

Graphical Abstract



Introduction

As scientific inquiry progressed, thin films have emerged as materials with distinctive properties which significantly differ from their bulk counterparts. Over the past few years, the thorough understanding and modification of thin film properties has had a significant impact on their use in various electronic applications. Scientists have invested substantial resources in the synthesis of semiconducting thin films. The combination of elements from Group II (metals) and Group VI (chalcogens) on the periodic table [1] produces compounds that exhibit semiconductor properties. These semiconducting materials have generated a great deal of interest in both fundamental research and practical technological applications [2]. Among these compounds, those derived from Group IIB and Group VI elements, such as zinc based chalcogenides (ZnO, ZnS, ZnSe, and ZnTe) and cadmium based chalcogenides (CdO, CdS, CdSe, and CdTe), have been widely used in an array of optoelectronic devices such as radiation detectors, solar cells, magneto-optical devices, visible light photodetectors, LEDs, magnetooptical devices, nonlinear optical materials, and others [3-14].

Zinc telluride (ZnTe) stands out as a leading material in this category of semiconductor compounds. The transparency of this p-type binary compound semiconductor to photons with energy levels below 2.26eV makes it an attractive choice for back contact with thin film solar cells [15-16]. ZnTe as a back contact in thin solar cells is aimed to enhance the overall performance of the solar cell by facilitating better light absorption and electron-hole pair generation within the thin film based solar cell configuration [17]. ZnTe is favoured for application in light-emitting diodes (LEDs) due to its advantageous direct band gap energy [18-20].

Furthermore, the band gap of ZnTe had been tuned by metal and non-metal ion doping such as Al [20-21], Sn [18,22-23], Cu [24-26], Sb [18,22,27-28], Bi [29], As [30], Cd [31-32], N [33], and P [34-35]. This possibility of tuning by dopants positioned ZnTe as potential candidate for various industrial applications. ZnTe thin films have been deposited using various techniques such as thermal evaporation [23,29,36-37], close space

sublimation (CSS) [38-39], glancing angle technique [40], chemical bath [19, 41-42], electrodeposition [43-47], pulsed laser deposition [48-49], reactive radiofrequency [33], metalorganic chemical vapor deposition (MOCVD) [30], and radio-frequency magnetron sputtering [50-51].

Electrochemical deposition, also known as electrodeposition or electroplating, is a versatile and widely used technique for depositing thin films and coatings onto conductive substrates. It involves the reduction of metal ions from a solution onto a conductive surface under the influence of an electric field [52]. This deposition technique presents several benefits, such as precise management of film characteristics, the ability to coat conformally, fast deposition rates, costeffective, suitability for multicomponent setups, eco-friendliness and flexibility [53-54]. These factors make it a preferred choice for thin film fabrication in various fields, ranging from microelectronics and optoelectronics to corrosion protection and decorative coatings. Many researchers such as [55-62] have reported the use of electrodeposition in the synthesis of binary, ternary, and quaternary thin films.

The motivation for the work arose from the fact that there is no literature available on the impact of deposition potential on the structural and optical properties of electrodeposited antimony doped zinc telluride (Sb:ZnTe) thin films. Reports from literatures on antimony doped zinc telluride thin films as presented by [18,22,27-28] showed no information on the effect of deposition potential on the optical and structural properties of Sb:ZnTe thin films. The purpose of this research is to address this literature gap by investigating the effect of deposition potential on the structural and optical properties of Sb:ZnTe thin films synthesized via electrodeposition. Specifically,

we aim to elucidate how different deposition potentials influence the structural and optical properties of electrodeposited Sb:ZnTe thin films.

Experimental

Reagents

The electrodeposition of antimony-doped zinc telluride (Sb:ZnTe) involved the use of antimony trichloride (SbCl₃) and zinc (II) acetate dihydrate (Zn(CH₃CO₂)₂·2H₂O) as precursors for antimony and zinc, respectively. Tellurium dioxide (TeO₂) served as the precursor for telluride ions, sodium sulfate acted as the supporting electrolyte, H₂SO₄ was used as pH adjuster and distilled water was utilized as the solvent medium.

Apparatus

The experimental setup utilized various apparatus, including 100 mL beakers for solution mixing, fluorine-doped tin oxide (FTO) glass substrates as working electrodes, an electronic compact scale for precise reagent measurements, and a magnetic stirrer hotplate for solution agitation. The electric energy was delivered by a DC power supply while the potential and current of the deposition were measured using digital multimeters. The reference electrode was an Ag/AgCl electrode, while the counter electrode was a platinum rod. Substrate degreasing was accomplished using an ultrasonic bath, and drying during the post-deposition heat treatment was made easier by an electrical oven.

Material preparation

Molar solutions of the reagents prepared for the deposition include:

(1) Zinc (II) acetate dihydrate solution (0.20M): Dissolve 4.39 grams of zinc (II) acetate dihydrate in 100 milliliters of distilled water.

(2) Antimony trichloride solution (0.05 M): Dissolve 1.14 grams of antimony trichloride in 100 milliliters of distilled water.

(4) Sodium sulphate (Na₂SO₄) solution (0.05 M): Dissolve 8.06 grams of sodium sulphate in 500 milliliters of double-distilled water.

(5) Zinc (II) acetate dihydrate, antimony trichloride, tellurium dioxide aqueous solutions were used as precursors for Zn, Sb, and Te ions while sodium sulphate was employed as a supporting electrolyte. In addition, the pH of the electrolytic bath was adjusted using $1.0 \text{ M} \text{ H}_2\text{SO}_4$.

Substrate pre-treatment

Four distinct processes were employed for the cleaning of fluorine-doped tin oxide (FTO) glass substrates before used in electrodeposition of films. These four processes are depicted in Figure 1.

Electrosynthesis of antimony doped zZinc tellurium thin films

Figure 2 illustrates the electrodeposition setup, which is made-up of the electrolyte, electrodes, and power supply unit. The electrodeposition setup of [63] was employed in this research work. As shown in Figure 2, the configuration involved a three-electrode electrodeposition setup which are working, reference and counter electrodes.

The working electrode, serving as the cathode, was the conducting substrate (FTO). A platinum electrode was used as the anode (counter electrode). A reference electrode of Ag/AgCl was utilized in the deposition of the thin film. The electrodeposition setup was powered by the Dazheng digital DC-power supply unit (PS-1502A) model. Two digital multimeters (DT9201A CE and Mastech MY60) were employed to measure deposition potential and current, respectively.

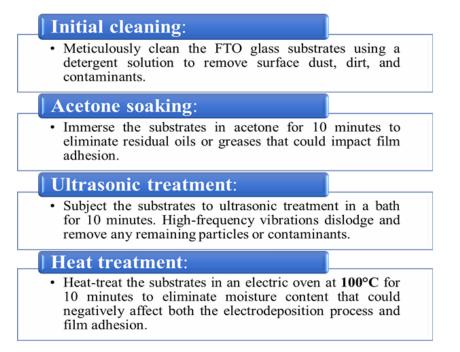


Figure 1. Pre-treatment steps for cleaning of FTO achieving optimal film quality.

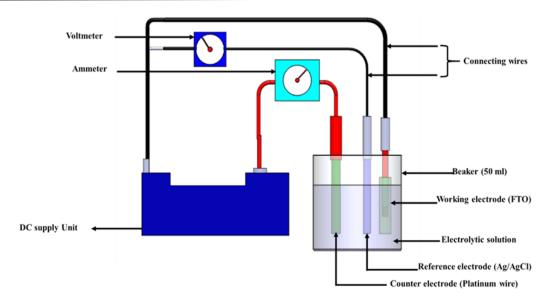


Figure 2. Schematic diagram of the electrodeposition experimental set up [63].

To deposit antimony doped zinc telluride thin films onto a FTO substrate using electrodeposition, a homogeneous aqueous electrolytic bath was formed by mixing 15 mL of 0.20 M zinc (II) acetate dihydrate. 10 mL of 0.05 M of antimony trichloride and 5 mL of concentrated 1 M of H₂SO₄. The mixture was properly mixed using a magnetic stirrer for a duration of 5 minutes. This follows with the addition of 15 mL of 0.10 M tellurium dioxide and 5 mL of 0.05 M Na₂SO₄. The resultant solution was employed after five more minutes of magnetic stirring. After that, the three electrodes were submerged in the electrolytic bath and kept at a steady 2.5-volt potential for precisely sixty seconds. To improve the characteristics of the deposited Sb:ZnTe thin film, the deposited layer was then thermally treated for 20 minutes at 100 °C. At various deposition potentials of 3.0, 3.5, 4.0, and 4.5 volts, four additional antimony-doped zinc telluride thin films were deposited. The mechanism of the formation of antimony doped zinc telluride is shown in Equation (1).

$$Zn^{2+} + Te^{4+} + Sb^{3+} + 2e^- \longrightarrow ZnTe:Sb \quad (1)$$

At the cathode (working electrode), Zinc ions (Zn^{2+}) , antimony ions (Sb^{3+}) , and tellurium ions (Te^{4+}) were reduced through electron transfer which lead to the deposition of the Zn, Sb, and Te metals. The simultaneous reduction of zinc, tellurium, and antimony ions results in the formation of the compound Sb:ZnTe on the electrode surface.

Characterization of deposited superlattice

The deposited antimony doped zinc telluride thin films were subjected to film thickness measurement using gravimetric method. Optical, electrical, and structural properties were also studied. Optical properties were carried out using 756S UV-Vis spectrophotometer. Structural analysis of the films was obtained using Drawell x-ray diffractometer.

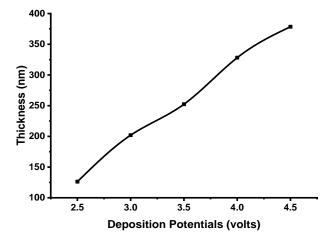


Figure 3. Graph of thickness against deposition potential.

Results and Discussions

Film thickness measurement

The thickness of the film was determined using the mass difference method, as described in Equation (2) in references [65-66].

(t)
$$=\frac{\Delta m}{\rho A'}$$
 (2)

Where, A is the area of the substrate covered by the deposited thin films, ρ is the bulk density of ZnTe (assumed to be 6.34 g/cm³), and Δ m is the mass difference found by measuring the substrate mass before and after the deposition.

A graph of the thickness of Sb-doped ZnTe film plotted against the deposition potential is presented in Figure 3. As the deposition potentials rose from 2.5 volts to 4.5 volts, the film thickness went up from 126.18 nm to 378 nm. Controlling the electrodeposition potential is crucial for achieving the desired film thickness and properties. As the deposition potentials increases, faster reduction rates are experienced by the ions resulting in thicker films because more metal ions are being reduced and deposited. This result is similar to the increase in thickness as deposition potential increases as obtained by [67-68].

Structural analysis

In Figure 4, the x-ray diffraction spectra of the thin films composed of Sb-doped ZnTe deposited on electrodes are presented. The diffractograms illustrate an x-ray pattern consistent with the hexagonal phase of ZnTe, as indicated by the JCPDS file number (00-019-1482). Structural phase of ZnTe obtained in this work is similar to those obtained by [69-70]. Equation (4) demonstrates how the Debye–Scherrer formula was used to calculate the crystallite sizes of the thin films that were deposited [71-72].

$$D = \frac{0.9\,\lambda}{\beta\cos\theta} \tag{3}$$

Dislocation densities of the deposited Sb doped ZnTe were calculated using Equation (4) as given by [73-74].

$$\delta = \frac{1}{D^2} \tag{4}$$

Microstrains of the deposited Sb doped ZnTe were calculated using Equation (5), respectively, as given by [75-76].

$$\varepsilon = \frac{\beta}{4\tan\theta} \tag{5}$$

Table 1 indicates the structural parameters of the Sb-doped ZnTe thin films that were deposited. In addition, Figures 5(a-c) illustrates the relationship between deposition potential and key structural characteristics, namely crystallite size (d), dislocation density (δ), and microstrain (ε). The crystallite size of the films ranges from 23.88 nm to 33.00 nm. The increase in crystallite size, as shown in Figure 5(a) often indicates improved structural phase. Larger crystallites generally have fewer grain boundaries. The increased crystalline structure of the films as a result of the higher deposition potential is responsible for the observed heightened intensity. Figures 5 (b and c) demonstrate that when deposition potential increases, dislocation density, and microstrain trend downward. A decrease in microstrain and dislocation density points to a reduced density of crystal defects. A more wellorganized and flawless crystal lattice is facilitated by fewer dislocations and reduced microstrain.

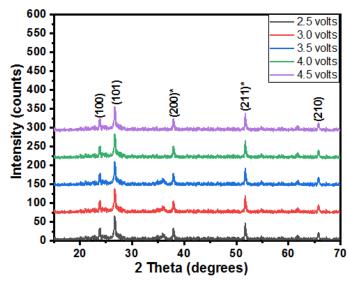


Figure 4. Diffractograms of antimony doped zinc telluride thin films deposited at varying potentials.

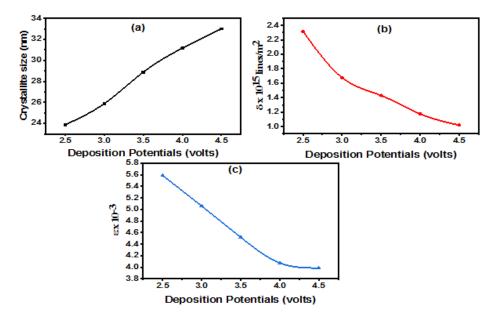


Figure 5. Variation of structural parameters (a) crystallite size (D), (b) dislocation density (δ) and microstrain (ε) with deposition potentials.

Table 1. Structural parameters of electrodeposited antimony doped zinc telluride thin film					
2θ(°)	d – spacing (nm)	FWHM (°)	D (nm)	$\delta x 10^{15}$	$\varepsilon x 10^{-3}$
				Lines/m ²	
23.255	3.822	0.543	15.595	4.112	11.518
26.715	3.334	0.315	27.046	1.367	5.794
37.943	2.369	0.302	29.071	1.183	3.831
51.732	1.766	0.291	31.728	0.993	2.616
65.831	1.418	0.620	15.939	3.936	4.180
			23.876	2.318	5.588
23.823	3.732	0.391	21.46	2.172	8.175
26.715	3.334	0.408	20.91	2.287	7.495
37.943	2.369	0.395	22.22	2.026	5.013
51.732	1.766	0.285	32.39	0.953	2.562
65.775	1.419	0.305	32.43	0.951	2.056
			25.92	1.678	5.060
23.873	3.724	0.265	31.98	0.978	5.474
26.765	3.328	0.478	17.85	3.139	8.764
37.993	2.366	0.293	29.75	1.13	3.738
51.782	1.764	0.285	32.39	0.953	2.560
65.825	1.418	0.305	32.44	0.95	2.054
			28.95	1.43	4.518
23.847	3.728	0.248	34.16	0.857	5.129
26.739	3.331	0.412	20.70	2.334	7.564
37.967	2.368	0.238	36.88	0.735	3.018
51.756	1.765	0.287	32.10	0.97	2.584
65.799	1.418	0.309	31.98	0.978	2.084
			31.17	1.175	4.076
23.263	3.821	0.325	26.09	1.469	6.883
26.715	3.334	0.305	27.97	1.278	5.603
37.944	2.369	0.287	30.52	1.073	3.648
51.733	1.766	0.255	36.1	0.767	2.299
65.775	1.419	0.223	44.32	0.509	1.504
		0.220	33.00	1.019	3.987

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Optical analysis

UV-Vis spectroscopy was employed to analyse the optical properties of electrodeposited thin films of antimony-doped zinc telluride (ZnTe) across a wavelength range of 400 nm to 1100 nm. The choice of the wavelength is because this range corresponds to the visible and nearinfrared (NIR) regions of the electromagnetic spectrum. As depicted in Figure 6, the absorbance of the antimony- doped ZnTe thin films exhibits a decline as the wavelength increases from 400 nm to 1100 nm. Moreover, a rise in absorbance is observed with an increase in the deposition potential from 2.5 volts to 4.5 volts. The absorbance values were observed to vary between 33% and 54%.

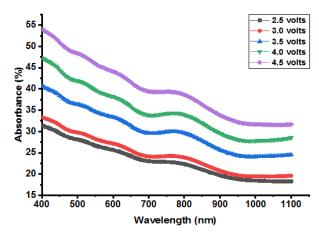


Figure 6. Absorbance spectra as a function of wavelength for antimony-doped zinc telluride (Sb:ZnTe) thin films at varied deposition potentials.

The graph in Figure 7 depicts transmittance spectra plotted against wavelength. As the wavelength increases from 400 nm to 1100 nm, the transmittance values of the films also increase. However, when the deposition potential increases, the transmittance decreases. These films exhibit moderate transmittance within the visible (VIS) region and high transmittance within the nearinfrared (NIR) region, with transmittance values ranging from 28.50% to 48.0%.

Figure 8 depicts the reflectance spectra of the films plotted against wavelength from 400 nm to 1100 nm. The reflectance values of the deposited thin films were consistently lower than 20.5%. The spectra illustrate a continuous decrease in reflectance as the wavelength increases for deposition potentials of 2.5 volts and 3.0 volts. With increasing deposition potential, reflectance is observed to rise within the VIS region and decline within the NIR region. Minimal reflectance values were observed within the NIR regions, indicating suitability for antireflective coating in smart window applications [65].

Figure 9 presents a graph of refractive index plotted against wavelength at different deposition potentials. The refractive indices of the films fall within the range of 2.40 to 2.63. Interestingly, the refractive index values decrease as the wavelength increases. However, when the deposition potential varies from 2.5 to 3.0 volts, the refractive index increases, and then it decreases again from 3.5 volts to 4.5 volts.

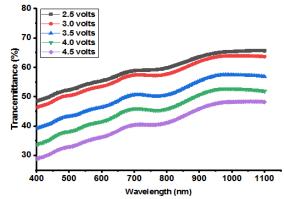


Figure 7. Transmittance spectra as a function of wavelength for antimony-doped zinc telluride (Sb:ZnTe) thin films at varied deposition potentials.

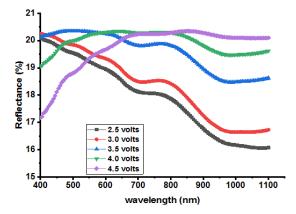


Figure 8. Reflectance spectra as a function of wavelength for antimony-doped zinc telluride (Sb:ZnTe) thin films at varied deposition potentials.

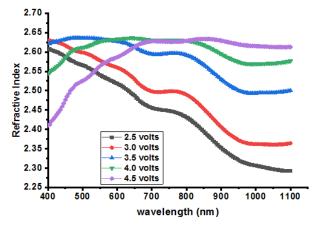


Figure 9. Refractive index as a function of wavelength for antimony-doped zinc telluride (Sb:ZnTe) thin films at varied deposition potentials.

Figure 10 displays the graph depicting the extinction coefficient (k) of antimony-doped zinc telluride thin films. The values were observed to rise with increasing wavelength across the studied spectrum (400 nm to 1100 nm) and decrease with higher deposition potential. Within the visible (VIS) region (400 nm to 700 nm), k values of the thin films ranged from 1.85 to 1.03, indicating effective photon energy absorption. Specifically, at deposition potentials of 2.5 volts, 3.0 volts, 3.5 volts, 4 volts, and 4.5 volts, k values ranged from 1.85 to 2.93, 1.24 to 1.97, 1.21 to 1.96, 1.04 to 1.75, and 1.03 to 1.68, respectively. The k measures the light absorption strength at a given wavelength. The spectrum of extinction coefficient values verifies the suitability of these films for application as absorber layers in thin-film solar cells.

In Figure 11, the relationship between $(\alpha hv)^2$ and photon energy is demonstrated for antimony-doped zinc telluride (Sb:ZnTe) thin films deposited at different deposition potentials. By extrapolating the linear part of the graph along the photon energy axis, the energy band gaps of these films were estimated. Specifically, a band gap of 2.00 eV was obtained for antimony-doped zinc telluride thin film deposited at 2.5 volts. For films deposited at 3.0 volts, 3.5 volts, 4.0 volts and 4.5 volts, the band gaps were found to be 1.95 eV, 1.90 eV, 1.90 eV, and 1.85 eV,

respectively. These results indicate a decrease in the energy band gap of the films as the deposition potential increases, suggesting the possible adjustment of energy band gap of antimony-doped zinc telluride by varying the deposition potential. These findings suggest that as the deposition potential increases, there is a decrease in the energy band gap of the films, implying the possibility of adjusting the energy band gap of antimony-doped zinc telluride by varying the deposition potential.

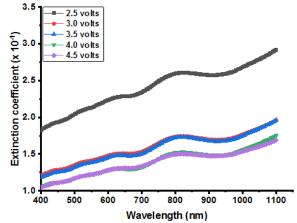


Figure 10. Extinction coefficient as a function of wavelength for antimony-doped zinc telluride (Sb:ZnTe) thin films at varied deposition potentials

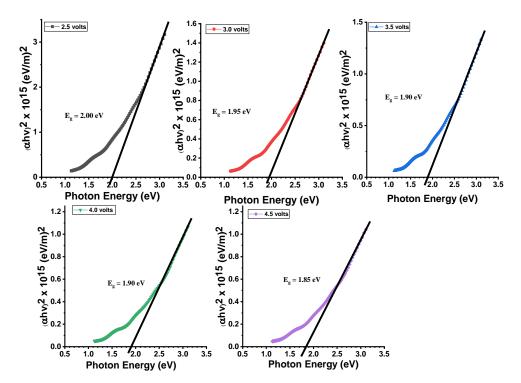


Figure 11. (αhv)² as a function of wavelength for antimony-doped zinc telluride (Sb:ZnTe) thin films at varied deposition potentials

These range of energy bandgap values positioned Sb:ZnTe thin films as potential materials for top junction (absorber layer) of a multijunction solar cells. The decrease in band gap of the Sb:ZnTe thin films as deposition potential increased from 2.5 volts to 4.5 volts could be attributed to improvement in the crystallinity and structural defects in the deposited films. These changes in grain size and crystallinity can impact the electronic band structure thereby modifying the band gap of the films. [75,77-79] have reported similar decrease in energy band gap of thin films as deposition potentials increases.

Conclusion

To sum up, the investigation into the electrodeposited antimony-doped zinc telluride (Sb:ZnTe) thin films has provided valuable insights into the influence of deposition potential on their structural and optical properties. The observed increase in film thickness, ranging from 126.18 nm to 378.55 nm, as the deposition potentials increased from 2.5 volts to 4.5 volts, underscores the critical role of control in achieving the desired film characteristics. Structural analysis done with X-ray diffraction, not only confirmed the hexagonal phase of ZnTe, but also revealed a substantial enhancement in crystallite size, ranging from 23.88 nm to 33.00 nm, and a simultaneous decrease in dislocation density and microstrain with higher deposition potential. The optical properties exhibited distinctive trends, with absorbance values ranging from 33% to 54%, transmittance from 28.50% to 48.0%. The energy band gap demonstrated tunability, decreasing from 2.00 eV at 2.5 volts to 1.85 eV at 4.5 volts.

The optical properties, characterized by tunable energy band gaps, absorbance values,

and high transmittance in the near-infrared region, indicate the potential suitability of these films for applications in solar energy harvesting. antireflective coatings, and transparent conductive coatings. The refractive indices spanning 2.40 to 2.63 further hint at their use in optical devices requiring controlled properties. These distinction refractive findings underscore the pivotal role of deposition potential in tailoring Sb:ZnTe thin films for specific applications, providing quantifiable insights into their optimization for thin-film solar cells, antireflective coatings, and the other optoelectronic devices.

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