

Comparative Study of Heat Treatment on the Optical Properties of Sb_2S_3 and CsCl thin Films at different Thicknesses

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ABSTRACT

A Sb_2S_3 and CsCl thin films has been prepared using thermal co-evaporation deposition technique. These films were annealed for different temperatures 100°C and 150°C . Absorbance and transmittance spectra were recorded in the wavelength range (300-900) nm. The nature of electronic transition were determined; it was found that this films has been indirect allowed transition with an optical energy gap. The extinction coefficient, refractive index, and energy bandgap was also measured before and after annealing.

Keywords : Sb_2S_3 and CsCl , Thin Film, Optical Properties.

I. INTRODUCTION

Recently antimony tri sulphide (Sb_2S_3) is useful material for various devices [1] optoelectronics, switching, television camera, and microwave. The use of Sb_2S_3 thin films in solar cell structural with conversion efficiencies of 5.5 and 7.3 % respectively has been reported by Nair & Pena [2]. In view of the various potential applications several methods of depositions [2, 3-22] has been employed to prepare thin films. Firstly cesium chloride (CsCl) nano crystals dispersed in antimony tri sulphide (Sb_2S_3) material of ratio 1 and 2. From the method used for the preparation of Sb_2S_3 and CsCl thin films, the thermal co-evaporation deposition method is often preferred because it offers large possibilities to modify the deposition condition so as to obtained films with determined structure and physical properties [9-22, 23] .the effect of heat treatment may promote chemical ordering after the molecular by a local rearrangement of atoms or change the number of defects in films [26]. In the present work we have examined the effect of heat treatment on CsCl nano crystal dispersed in antimony tri sulphide (Sb_2S_3) at different thickness. Thin films had been annealed at

different temperature i.e. 100°C and 150°C for one hour in air using horizontal muffle Furness. The optical and structural properties [24, 25] of thin film are changed after annealing the deposited films.

II. EXPERIMENTAL DETAILS

Thermal co-evaporation is the most commonly used technique for deposition of Sb_2S_3 and CsCl thin films on the glass substrate at room temperature by vacuum ($< 10^{-5}$). Evaporation using crystalline antimony tri sulphide and CsCl as a starting material in a molybdenum boat. The grown films were in amorphous nature. The choice of this method arises from its low cost, ease of handling and possibility of application on large surface of glass substrate were used. The films are prepared by taking equimolar solutions of Sb_2S_3 and CsCl in appropriate volumes in a ratio 2:1. Small pieces of small size $0.6 \times 0.6 \text{ cm}^2$ were cut from the bigger as grown samples. They were placed for annealing for one hour on a Horizontal Muffle Furnace in air at the desired temperatures 100°C and 150°C with an error of $\pm 5^\circ\text{C}$. The temperature at the top surface of the empty substrate reaches the temperature homogeneously of the Furnace in about

one hour. The optical constants of these films were determined through transmission and absorption measurements by the means of UV/VIS double beam spectrometer Shimadzu JAPAN (model UV-2401 PC shimadzu) [24]. The primary aim of the present study was to examine changes in optical properties of such films with temperature at different thicknesses.

III. RESULT AND DISCUSSION

3.1. Optical properties

The optical properties and solid state characterizations performed on the films grown on this work are the absorbance A, the transmittance T, and the band gap E_g of the Sb_2S_3 and CsCl thin films. The absorbance and transmittance characterization was carried out using a UV-2401 PC spectrometer in the visible and near infra red region for five different thickness films is displayed, but in present work we have show three graph one of them room temperature second 100°C and third 150°C shown in **figure 1 (a), (b) and (c)**. The refractive index (n) was determined as a function of wavelength by drawing a continuous curve on

maxima and minima of the interference fringe pattern of spectra as described in ref [25]. We could determine n for samples annealed at temperature $\leq 160^\circ\text{C}$, since the fringe pattern decreases in amplitude at higher temperatures.

Simplifying the refractive index is given as

$$n_f = \sqrt{N + \sqrt{N^2 - n_s^2}}$$

Where

$$N = 2n_s \frac{T_M - T_m}{T_M T_m} + \frac{n_s^2 + 1}{2}$$

The accuracy to which λ (wavelength) can be measured depends on the scale used and for the case of **figure 1 (a), (b) & (c)** the maximum accuracy of T_M and T_m is also about 0.001 or 0.1%. These two values set the limits for the accuracy of the calculated values of n and α (extinction coefficient) [27]. **Table-I, II & III** shows the values at the extremes of the spectrum of λ , T_M and T_m obtained from **figure.2** at different temperature.

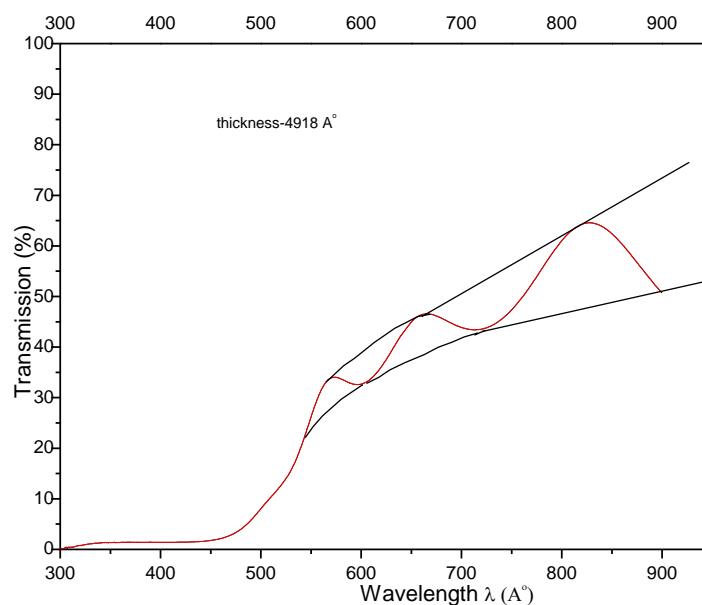


Figure 1 (a). Transmission spectra of Sb_2S_3 and CsCl thin film at room temperature

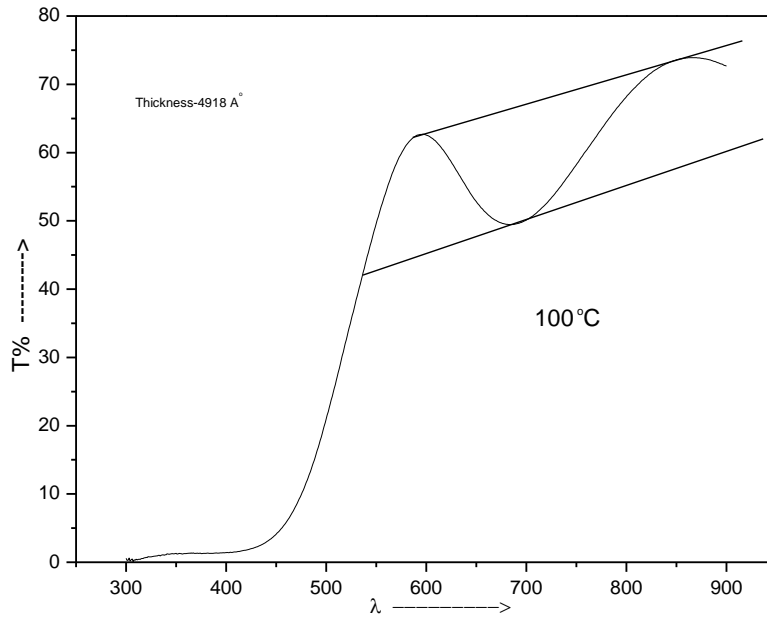


Figure 1 (b). Optical transmission spectra of Antimony Tri-Sulphide (Sb_2S_3) and Cesium Chloride ($CsCl$) thin film treated at $100^\circ C$.

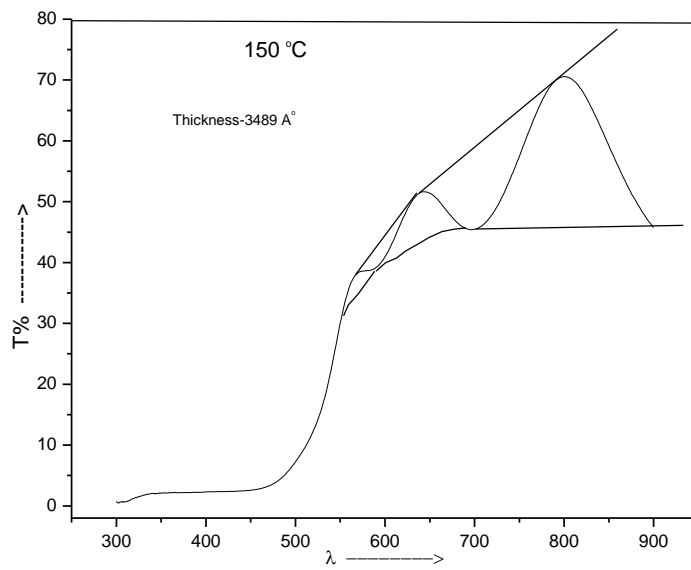


Figure 1 (c). Optical transmission spectra of Antimony Tri-Sulphide (Sb_2S_3) and Cesium Chloride ($CsCl$) thin films treated at $150^\circ C$.

Sample name	S.No.	λ (nm)	T_M %	T_m %	t (A ^o)	R.I.
S-12	1	600	66.5	56.5	2006	2.080123
	2	700	75.0	65.5	2006	1.954875
	3	800	79.0	69.0	2006	1.917314
S-13	4	600	38.4	29.2	2822	2.808503
	5	700	54.0	40.0	2822	2.609294
	6	800	61.6	47.2	2822	2.415796
S-14	7	600	43.2	31.6	3489	2.840709
	8	700	55.5	41.6	3489	2.552845
	9	800	61.6	49.6	3489	2.274078
S15	10	600	38.61	32.22	4918	2.440095
	11	700	51.387	41.665	4918	2.360204
	12	800	64.445	48.335	4918	2.444716
S-16	13	600	79.211	63.682	6062	2.146841
	14	700	86.575	71.315	6062	2.048623
	15	800	88.416	72.63	6062	2.046376

Table.I. Variation of refractive index (n) for a Sb₂S₃ and CsCl thin films with wavelength before annealing where λ is wavelength, T_M (upper extreme point) and T_m (lower extreme point), t is thickness and n is refractive index.

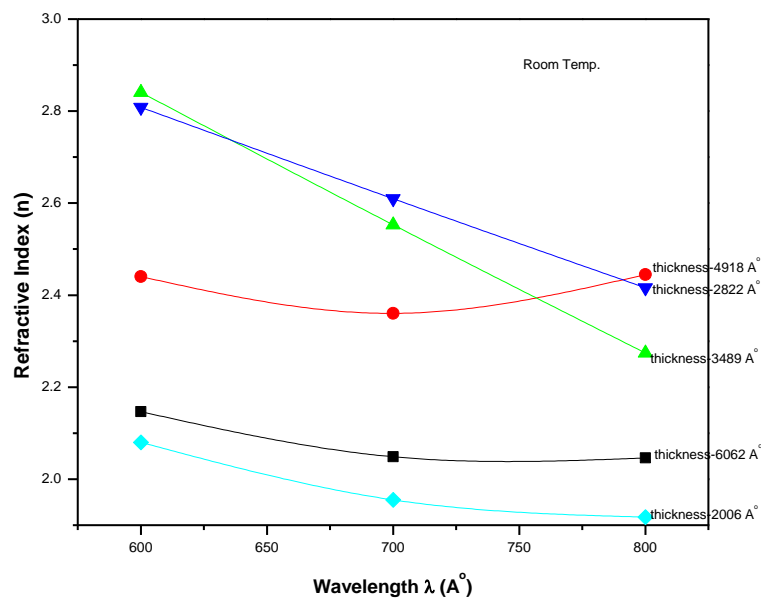


Figure 2. Graph between refractive index values of Antimony Tri-Sulphide (Sb₂S₃) and Cesium Chloride (CsCl) thin films with wavelength at room temperature

Sample name	S.No.	λ (nm)	T_M %	T_m %	t (Å ⁰)	R.I.
S-17	1	600	66.5	56.5	2006	1.633151
	2	700	75.0	65.5	2006	1.862881
	3	800	79.0	69.0	2006	1.831745
S-18	4	600	38.4	29.2	2822	2.829882
	5	700	54.0	40.0	2822	2.633014
	6	800	61.6	47.2	2822	2.47446
S-19	7	600	43.2	31.6	3489	2.610887
	8	700	55.5	41.6	3489	2.448978
	9	800	61.6	49.6	3489	2.324578
S-20	10	600	38.61	32.22	4918	2.748123
	11	700	51.387	41.665	4918	2.520191
	12	800	64.445	48.335	4918	2.422139
S-21	13	600	79.211	63.682	6062	2.564053
	14	700	86.575	71.315	6062	2.256388
	15	800	88.416	72.63	6062	2.093346

TableII. Variation of refractive index (n) for Sb₂S₃ and CsCl thin films with wavelength after annealing (100°C) where λ is wavelength, T_M (upper extreme point) and T_m (lower extreme point), t is thickness and n is refractive index.

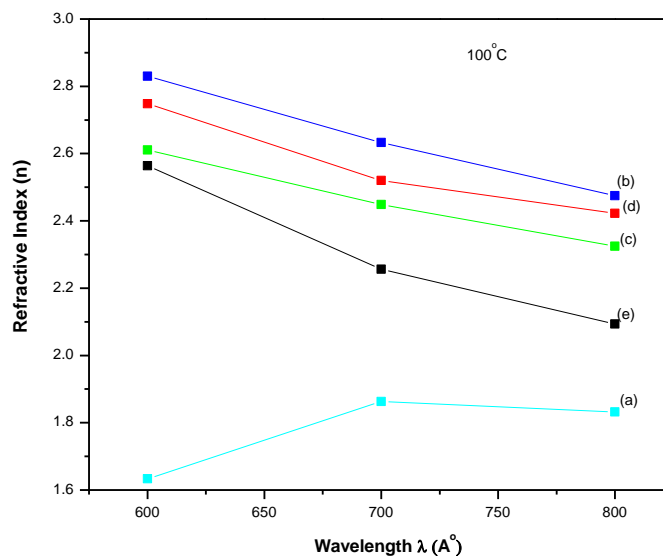


Figure 3. Variation of refractive index vs Wavelength at 100°C (a) - thickness 2006 Å (b)-thickness 2822 Å (c)- thickness 3489 Å (d)-thickness 4918 Å (e)-thickness 6062 Å.

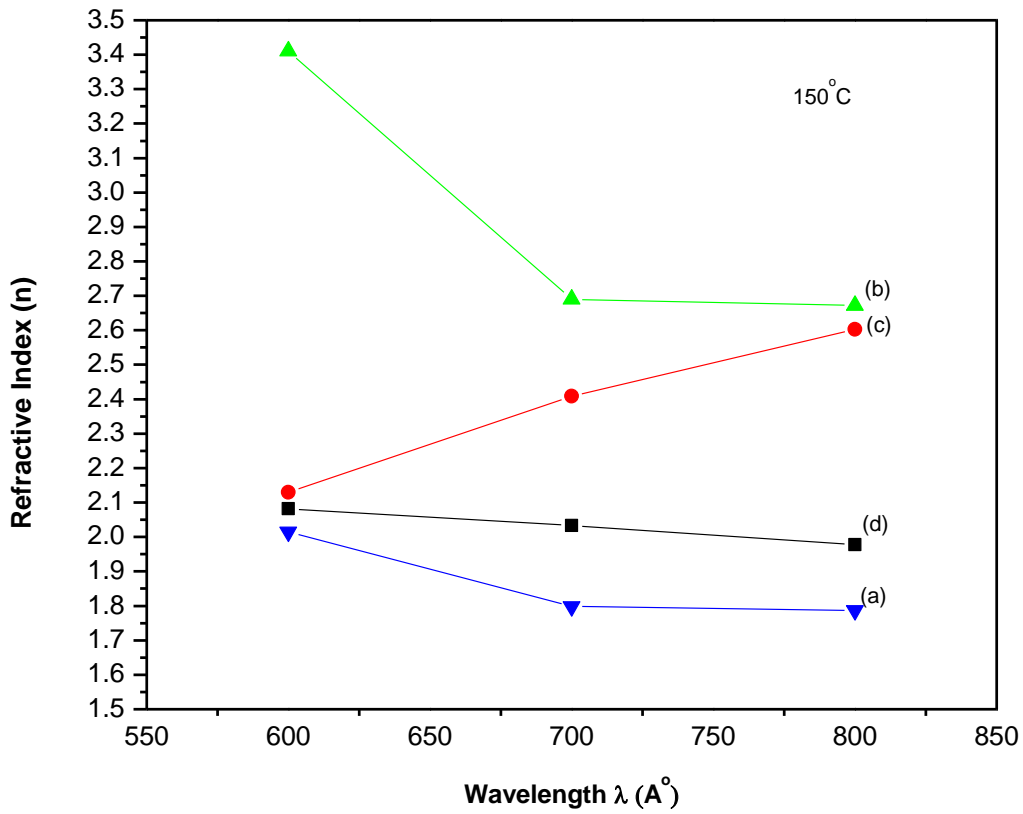


Figure 4. Variation of refractive index vs wavelength at 150°C (a) - thickness 2006 Å (b)-thickness 2822 Å (c)- thickness 3489 Å (d)-thickness 6062 Å

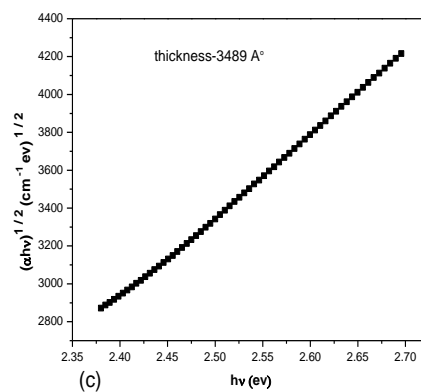
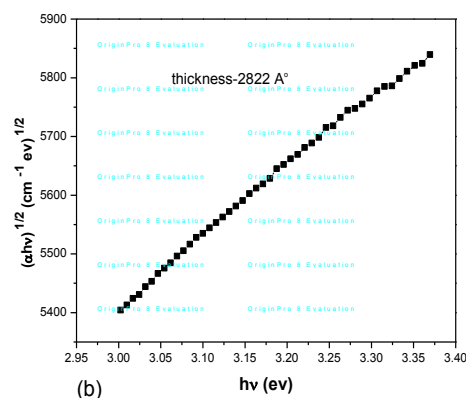
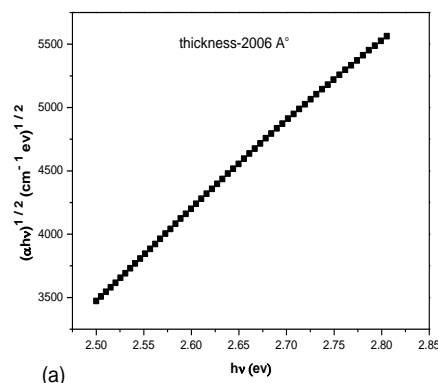
Sample name	S.No.	λ(nm)	T _M %	T _m %	t (Å ⁰)	R.I.
S-22	1	600	66.5	56.5	2006	2.015534
	2	700	75.0	65.5	2006	1.798001
	3	800	79.0	69.0	2006	1.787065
S-23	4	600	38.4	29.2	2822	3.409961
	5	700	54.0	40.0	2822	2.68989
	6	800	61.6	47.2	2822	2.671731
S-24	7	600	43.2	31.6	3489	2.129599
	8	700	55.5	41.6	3489	2.409222
	9	800	61.6	49.6	3489	2.602736
S-25	10	600	38.61	32.22	6062	2.08145
	11	700	51.387	41.665	6062	2.03297
	12	800	64.445	48.335	6062	1.97811

Table III. Variation of refractive index (n) for Sb₂S₃ and CsCl thin films with wavelength after annealing (150°C). Where λ is wavelength, T_M (upper extreme point) and T_m (lower extreme point), t is thickness and n is refractive index.

The optical absorption spectra of the as deposited and annealed films are recorded at room temperature, 100°C & 150°C are shown in **figure 1 (a), (b) & (c)**. The values of the optical absorption coefficient α were not corrected for the transmittance and the reflectance of the films surface. The optical data is analyzed from the following classical expression for near- edge optical absorption in semiconductors [27].

$$\alpha h\nu = k (h\nu - E_g)^{n/2}$$

Where k is excitation coefficient, E_g is the semiconductor band gap and n is a constant equal to 1 for direct band gap and 4 for indirect band gap. Since the graph plots $(\alpha h\nu)^{1/2}$ against $h\nu$ are linear, the indirect nature of the optical transitions in Sb₂S₃ and CsCl is confirmed. Where α is the optical absorption coefficient, A is a constant, $h\nu$ is the energy of the incident photon, E_g is the optical band gap and n is an index which could take different values according to the electronic transition. After we try all the possible values of n we conclude that kind of transition is direct which is in good agreement with Ubale et. al. [28], the characteristics of $(\alpha h\nu)^{1/2}$ vs $h\nu$ (photon energy) were plotted for evaluating the band gap (E_g) of the Sb₂S₃ thin films, and extrapolating the linear portion near the onset of absorption edge to the energy axis as shown in **figures (5), (7) (8)** shows the value of E_g before and after annealing, it can be noticed that the value of the optical energy gap decreases as the annealing temperature increases, this could be attributed to the fact that an increase in annealing temperature leads to minimizing structural imperfections in the as deposited thin films. This behavior was in good agreement with Versavel and Haber [29].



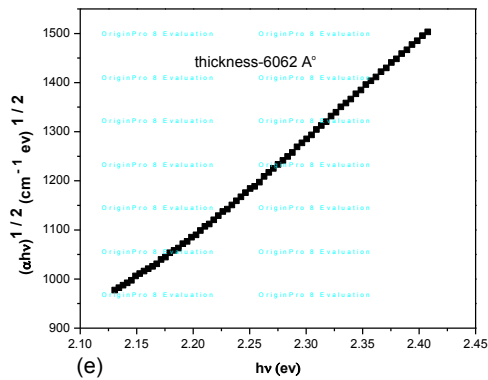
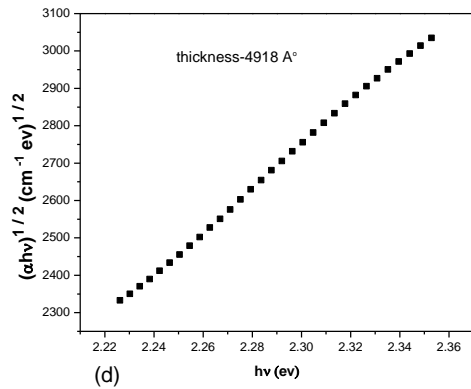


Figure 5. (a), (b), (c), (d) and (e) Plot $(\alpha hv)^{1/2}$ versus $h\nu$ for the direct band gap of Sb_2S_3 and $CsCl$ thin films before annealing.

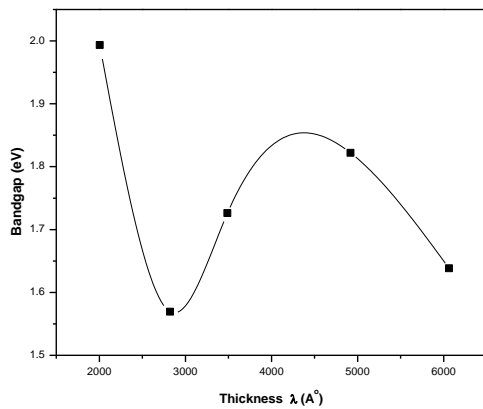


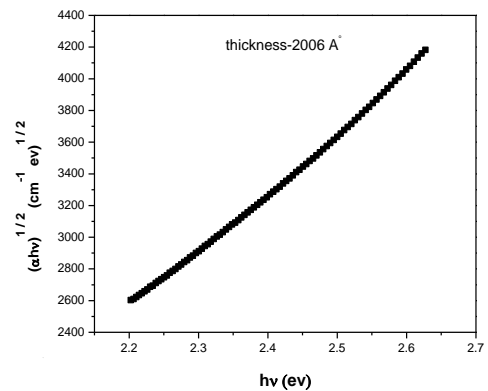
Figure 6. Plot graph between bandgap (eV) versus thicknesses (\AA) of Antimony Tri-Sulphide (Sb_2S_3) and Cesium Chloride ($CsCl$) thin films before annealing.

Extrapolation of these curves to zero absorption coefficients gives the optical energy gap E_g which is

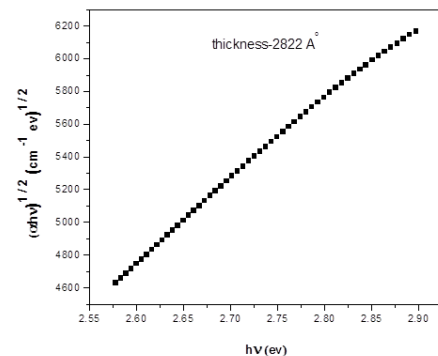
shown in Table IV before and after annealed. An annealed sample shows that there is decrease in E_g after heat treatment of as-deposited Sb_2S_3 and $CsCl$ films. Comparative study of energy bandgap (E_g) of deposited thin films before and after annealing is shown in figure 9.

S.No.	t (thickness)	BAND GAP (E_g)		
		Room Temp.	100°C	150°C
1	2006	1.993604	1.511	1.60971
2	2822	1.569439	1.627833	1.836903
3	3489	1.726239	1.950948	1.88336
4	4918	1.821932	1.902274	-
5	6062	1.638372	1.916446	1.331853

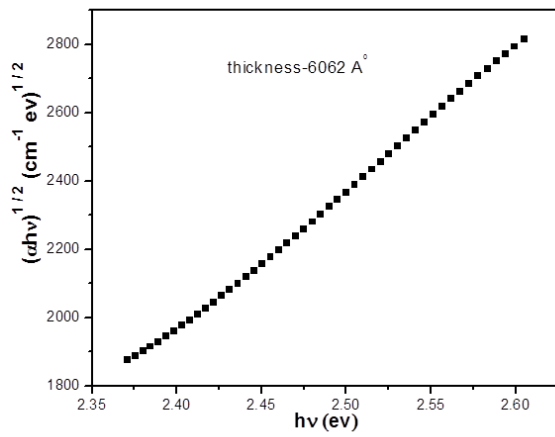
Table-IV. Bandgap (E_g) at different thickness before and after annealing.



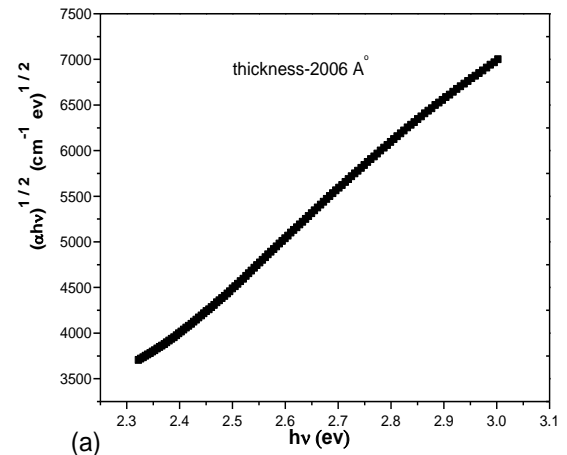
(a)



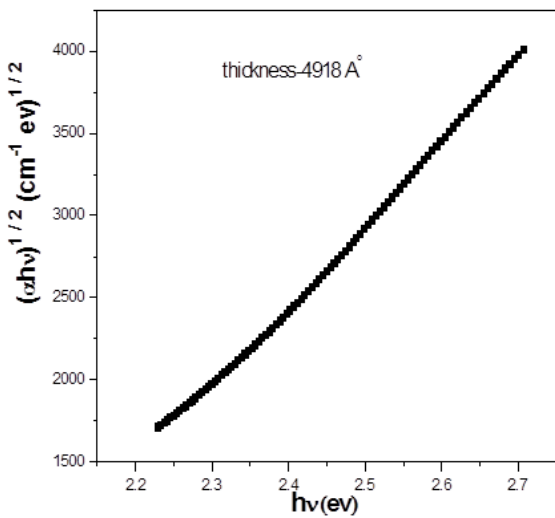
(b)



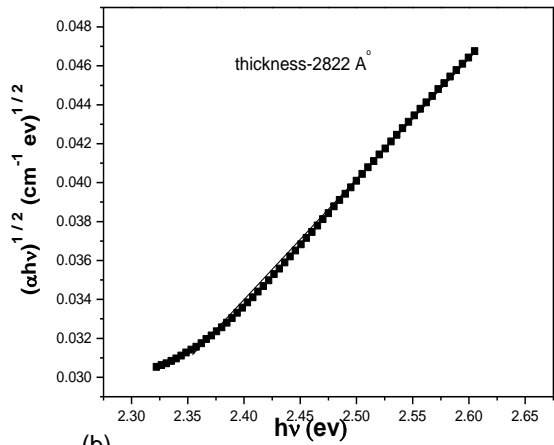
(c)



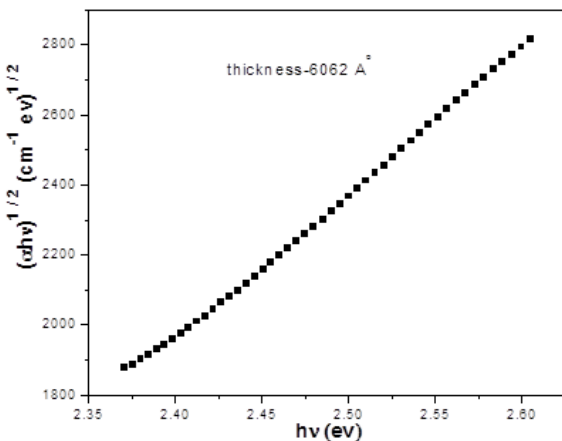
(a)



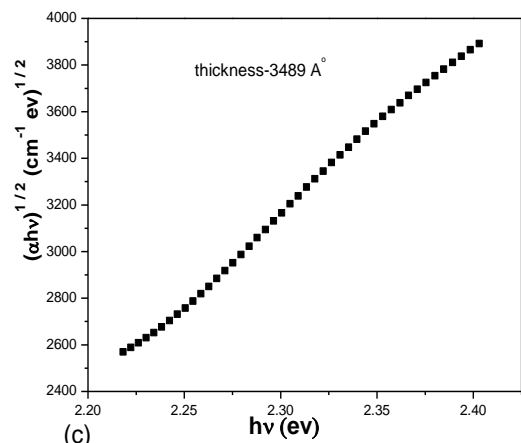
(d)



(b)



(e)



(c)

Figure 7. (a), (b), (c), (d) and (e) Plot of $(\alpha hv)^{1/2}$ versus $h\nu$ for Antimony Tri-Sulphide (Sb_2S_3) and Cesium Chloride ($CsCl$) thin film at temperature $100^\circ C$. The straight lines show the least square fitting used for determining E_g by extrapolation.

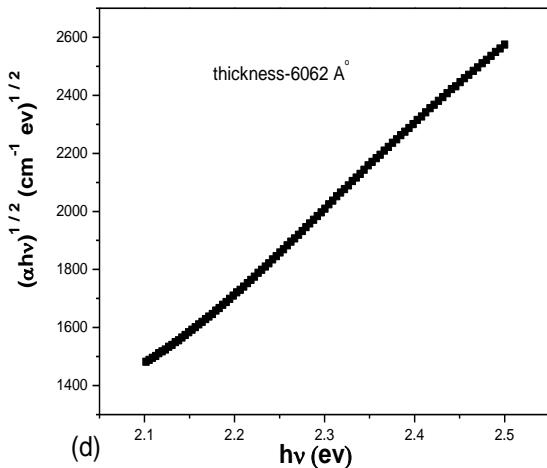


Figure 8. Plot of $(\alpha hv)^{1/2}$ versus hv for Antimony Tri-Sulphide (Sb_2S_3) and Cesium Chloride ($CsCl$) thin film at temperature $150^\circ C$. The straight lines show the least square fitting used for determining E_g by extrapolation.

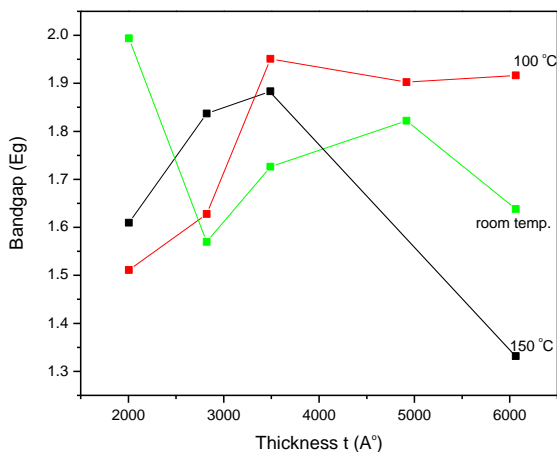


Figure 9. Comparative study of bandgap (E_g) vs. thickness of Sb_2S_3 and $CsCl$ deposited thin films before and after annealing.

IV. CONCLUSION

Thin films of Sb_2S_3 and $CsCl$ ratio 2:1 were successfully deposited using thermal co-evaporation technique of various thicknesses. Before and after annealing grown films of Sb_2S_3 and $CsCl$ are amorphous in nature. The refractive index varies from

1.9173 to 2.84 at room temperature and 1.63 to 2.83 at $100^\circ C$ and 1.787 to 3.40 at $150^\circ C$ at different wavelength and thicknesses. The films were found to be an indirect bandgap (E_g) 1.99ev, 1.56ev, 1.72ev, 1.82ev, and 1.63ev at room temperature and after annealing at $100^\circ C$ the bandgap (E_g) was 1.51ev, 1.62ev, 1.95ev, 1.90ev and 1.91ev and at $150^\circ C$ the bandgap (E_g) was of the films 1.60ev, 1.83ev, 1.88ev and 1.33ev. By the comparative study of bandgap before and after annealing, bandgap increases with thickness. Which explain the value of the optical energy gap was affected by annealing which was decreased as the annealing temperature increase the film deposited in this work shows narrow band gap, as such it could serve as good absorber layers for photocells.

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