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STUDIES ON STRUCTURAL AND OPTICAL PROPERTIES OF VACUUM EVAPORATED In_2Te_3 THIN FILMS

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In_2Te_3 thin films were grown on mica substrates by thermal evaporation method under high vacuum with quartz crystal thickness monitor used for measurement of thickness of film. Lattice parameters and particle size (D) has been calculated from XRD data. The particle size is found to be in the range of 22-68nm for In_2Te_3 thin films deposited on mica substrate. The optical constants absorption coefficient (α) , extinction coefficient (k), refractive index (n), dielectric constant (real and imaginary), absorption coefficient (α) and optical band gap (E_g) are reported for In_2Te_3 films deposited on mica substrates. The absorption coefficient (α) of In_2Te_3 thin films was found to be in order of 10^6 with refractive index (n=) 1.56.

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1. Introduction

Binary chalcogenide alloys $A_2^{III}B_3^{VI}$ (A=Al,Ga,In and B=S,Se,Te) with semiconducting properties have been widely studied during the last few decades. In_2Te_3 is an III-VI compound with a band gap of about 1.1 eV and is important for technical application in IR detectors, memory devices, switching, thermoelectric generator, gas sensors, strain gauges and solar cells. It crystallize in zinc blend structures with a lattice parameter a=6.16 A^0 with 1/3rd of Indium sites are vacant. It also crystallize in α-phase as a super lattice with a=18.54 A^0 of amorphous films. It exhibits negative temperature coefficient and have very high electrical resistivity at room temperature. Roughieh Rousina and G.K.Shiva Kumar [1, 2] studied the growth and characterization of Indium Telluride thin films grown by conventional vacuum evaporation technique. They made stoichiometric films by non-Stoichiometric bulk was used, they found the bulk composition In: Te = 55.7: 44.3 yields stoichiometric films. Indium deficient films were obtained on annealing at elevated temperatures. It crystallizes TIS type structure. I-V characteristics are also studied as grown indium telluride with metal contacts made schottky diodes. P.C. Mathur et al [3] studied the growth of Indium Telluride thin films on sapphire (Al_2O_3) substrates by thermal evaporation at 350 ^{0}C and annealed in an argon atmosphere at 400 ^{0}C for 2 hours. D.C. conductivity and Hall Effect studies were carried out in the temperature range 77 – 295 K. The Hall coefficient ,Hall mobility, Band gap and carrier concentration were found to be 5 X 10^5 cm^3 C^{-1},112 cm^2 $V^{-1}S^{-1}$,1.02 eV ,7.7 X 10^{13} cm^{-3} respectively.

Electron beam evaporated indium telluride thin films were found optimum substrate temperature T_0 of 453 K observed by S.De Purkayastha et al [4]. They exhibit maximum hole mobility of 48 cm^2 V^{-1} S^{-1} and maximum hole concentration of 4.2 X 10^{18} cm^{-3} and explained on the basis of the Vincett – Barlow – Robers theory. The activation energies were found to be 0.039

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eV below 300 K and 0.12 eV above 300 K. Hall coefficients and d.c. conductivity studies in the temperature range 100-625 K for polycrystalline In_2Te_3 films grown on glass and mica substrates structural change was observed at higher temperatures by P.C. Mathur et al [5] The α-phase transforms to the β-phase at a temperature of above 620 ^{0}C.

Structural and electrical characterization of Indium Telluride thin films were carried by Roughieh Rousina and G.K.ShivaKumar [6]. They found the activation energies are 0.45 eV and 0.96 eV for low and high temperature regions respectively. The switching phenomenon in amorphous In_2Te_3 thin films was studied by M.A.Afifi et al [8]. The switching voltage V_{th} was linear with thickness and V_{th} decreases exponential with temperature rise from 298 – 373 K. The switching voltage activation energy with temperature dependence of V_{th} is about 0.25 eV. The conduction activation energy was found to be 0.51 eV and the switching phenomenon in In_2Te_3 will be explained according to an electro thermal model. N.A.Hegab et al [9, 10] studied effect of annealing on the structural and electrical properties of In_2Te_3 deposited by thermal evaporation on glass substrate and found to be amorphous. The electrical conductivity studies reveal the activation energies are found to be 0.521eV and the corresponding room temperature electrical conductivity is 7.15 X 10^{-7} $\Omega^{-1}m^{-1}$.

In the present work, we studied the structural and optical properties of In_2Te_3 thin films prepared by thermal evaporation technique. The optical band gap, refractive index, extinction coefficient and dielectric properties are significantly dependent on the film thickness.

2. Experimental details

In_2Te_3 compound was synthesized first, by mixing and melting the high purity (99.999%) individual elements in stoichiometric proportions in a sealed quartz ampoule. The resultant bulk was then used as the source materials for the deposition of In_2Te_3 thin films. In_2Te_3 thin films were deposited in vacuum of 10^{-5} Torr on mica substrates at different substrate temperatures and thicknesses by the thermal evaporation technique using vacuum coating unit model 12A4 DM (Hind Hivac Company, Bangalore, India). XRD patterns of the films were recorded with the help of Philips (PW 1830) X-ray diffractometer using CuKα radiation. The tube was operated at 30KV, 20mA with the scanning speed of 0.030(2θ)/sec. The normal incidence transmission spectra of thin films of the samples have been measured by using Perkin-Elmer lamda-19, double beam spectrophotometer in the range 200 – 2000 nm.

3. Results and discussion

The prepared material was analyzed by X-ray diffraction and was found to have cubic structure with lattice constant a = 6.045A^0. Particle size (D) was estimated from X-ray diffraction data using Scherer formula [11]

$$D= (0.94\lambda)/(\beta \cos \theta) \quad (1)$$

where λ (=1.54A^o) is source wavelength, β is full width half maximum (FWHM), θ is peak position and found to be in the range of 22-68nm for In_2Te_3 thin films deposited on mica substrate. It is observed that the particle size increases with increase in thickness and substrate temperatures of In_2Te_3 films. The peaks have been indexed with the help of a computer program – POWDIN [12] using the observed interplanar spacing d.

The lattice constant 'a' for the cubic phase structure is determined by the relation

$$a = d/(h^2+k^2+l^2)^{1/2} \quad (2)$$

Strain values ε, can be evaluated by using the following relation [13]

$$\varepsilon = \beta\cos\theta/4 \quad (3)$$

Dislocation density (δ) has been calculated by using the formula [14]

$$\delta = 15\varepsilon/aD \quad (4)$$

Fig. 1 shows the Intensity versus 2θ of In_2Te_3 thin films of various thicknesses deposited on mica substrates at 303K and 353K. The XRD spectra exhibit position of the peak shifts towards higher angles. Tables 1-4 shows the observed and calculated values using Powdin program for X-ray diffraction data of In_2Te_3 thin films of various thicknesses deposited on mica substrates at 303K and 353K. The various structural parameters for In_2Te_3 thin films deposited at different substrate temperatures are calculated using the relevant formulas and are systematically represented in Table 5.

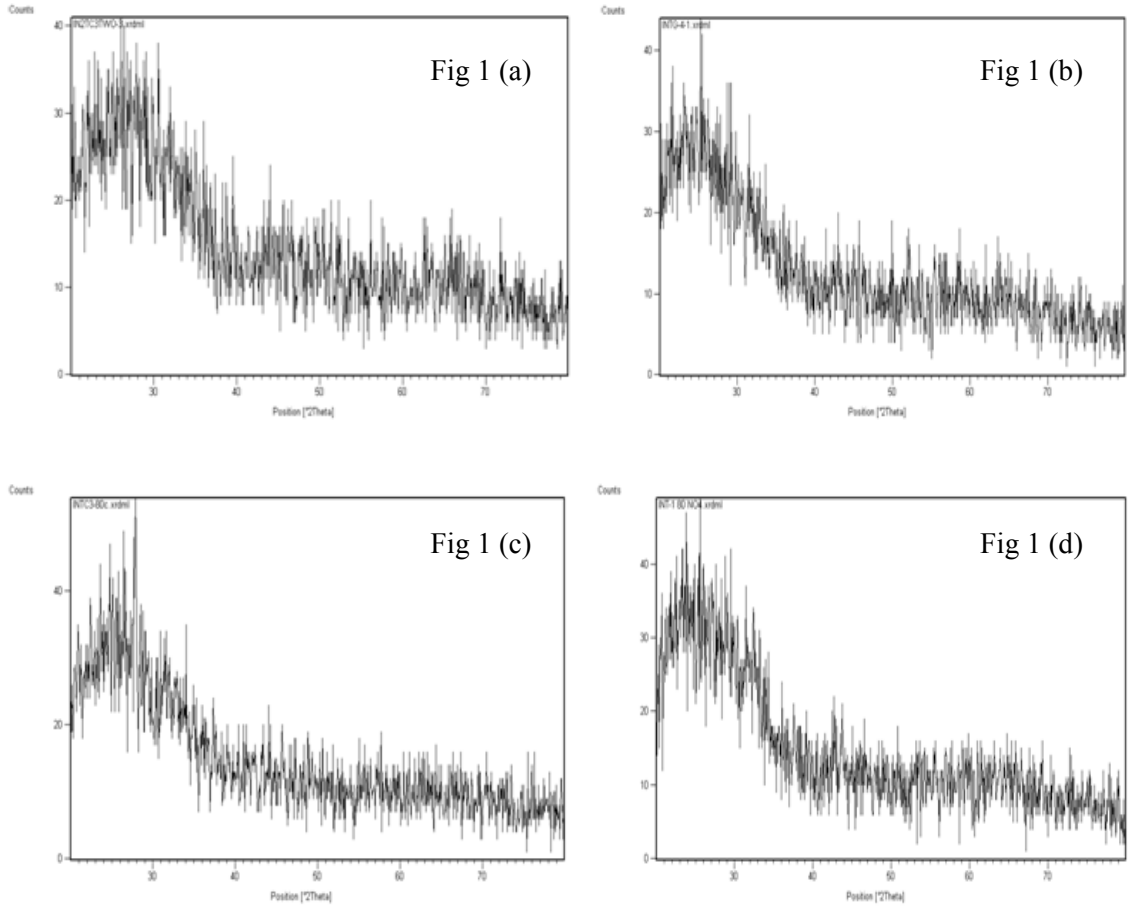


Fig 1. XRD patterns for In_2Te_3 thin films deposited on mica substrates of various thickness (a: 1000A^o, b: 1600A^o, c: 600A^o, d: 800A^o)

Table. 1. Observed and Calculated values using Powdin program for X-ray diffraction data of In_2Te_3 thin films with thickness 800A^o deposited at 303K temperature.

Line		d-spacing A.		Indices			SinSqTheta*E4		2Theta Deg.		
o.	c.	obs.	calc.	h	k	l	obs.	calc.	obs.	calc.	diff
1	1	3.5014	3.5565	1	1	1	483.9	469.1	25.42	25.02	.400
2	2	3.0002	3.0800	2	0	0	659.1	625.4	29.75	28.96	.788
3	3	2.1795	2.1779	2	2	0	1249.0	1250.8	41.39	41.42	-.032
4	4	1.9311	1.8573	3	1	1	1591.0	1719.9	47.01	49.00	-1.988
5	5	1.7575	1.7782	2	2	2	1920.8	1876.2	51.99	51.34	.651
6	6	1.5763	1.5400	4	0	0	2387.8	2501.7	58.50	60.02	-1.518
7	7	1.4614	1.4132	3	3	1	2778.0	2970.7	63.61	66.06	-2.440
8	8	1.3705	1.3774	4	2	0	3158.7	3127.1	68.39	68.00	.391
9	9	1.2604	1.2574	4	2	2	3734.7	3752.5	75.34	75.55	-.211
10	10	1.2062	1.1855	5	1	1	4077.8	4221.5	79.37	81.04	-1.671

Table. 2. Observed and Calculated values using Powdin program for X-ray diffraction data of In_2Te_3 thin films with thickness 1600A^o deposited at 303K temperature.

Line		d-spacing A.		Indices			SinSqTheta*E4		2Theta Deg.		
o.	c.	obs.	calc.	h	k	l	obs.	calc.	obs.	calc.	diff
1	2	3.1000	3.0450	2	0	0	617.4	639.9	28.77	29.31	-.531
2	3	2.1295	2.1531	2	2	0	1308.3	1279.7	42.41	41.92	.488
3	4	1.8445	1.8362	3	1	1	1743.9	1759.7	49.37	49.60	-.238
4	5	1.7774	1.7580	2	2	2	1878.0	1919.6	51.36	51.97	-.608
5	6	1.5469	1.5225	4	0	0	2479.4	2559.5	59.73	60.78	-1.057
7	8	1.3278	1.3618	4	2	0	3365.1	3199.4	70.91	68.89	2.023
8	9	1.2109	1.2431	4	2	2	4046.5	3839.2	79.01	76.58	2.430

Table. 3. Observed and Calculated values using Powdin program for X-ray diffraction data of In_2Te_3 thin films with thickness 600A^o deposited at 353K temperature.

Line		d-spacing A.		Indices			SinSqTheta*E4		2Theta Deg.		
o.	c.	obs.	calc.	h	k	l	obs.	calc.	obs.	calc.	diff
1	1	3.5959	3.5565	1	1	1	458.8	469.1	24.74	25.02	-.279
2	2	2.9882	3.0800	2	0	0	664.4	625.4	29.88	28.96	.910
3	3	2.1260	2.1779	2	2	0	1312.6	1250.8	42.48	41.42	1.059
4	4	1.8650	1.8573	3	1	1	1705.7	1719.9	48.79	49.00	-.215
5	5	1.6969	1.7782	2	2	2	2060.4	1876.2	53.99	51.34	2.655
6	7	1.4470	1.4132	3	3	1	2833.6	2970.7	64.32	66.06	-1.732
7	8	1.3520	1.3774	4	2	0	3245.8	3127.1	69.46	68.00	1.459
8	9	1.2684	1.2574	4	2	2	3687.7	3752.5	74.78	75.55	-.768
9	10	1.2034	1.1855	5	1	1	4096.8	4221.5	79.59	81.04	-1.450

Table. 4. Observed and Calculated values using Powdin program for X-ray diffraction data of In_2Te_3 thin films with thickness 1000A^o deposited at 353K temperature.

Line		d-spacing A.		Indices			SinSqTheta*E4		2Theta Deg.		
o.	c.	obs.	calc.	h	k	l	obs.	calc.	obs.	calc.	diff
1	3	2.1149	2.1779	2	2	0	1326.4	1250.8	42.72	41.42	1.293
2	4	1.8494	1.8573	3	1	1	1734.6	1719.9	49.23	49.00	. 223
3	5	1.7970	1.7782	2	2	2	1837.3	1876.2	50.76	51.34	-.574
4	6	1.5298	1.5400	4	0	0	2535.1	2501.7	60.46	60.02	.442
5	7	1.4557	1.4132	3	3	1	2799.8	2970.7	63.89	66.06	-2.162
6	8	1.3569	1.3774	4	2	0	3222.4	3127.1	69.17	68.00	1.173
7	9	1.2993	1.2574	4	2	2	3514.4	3752.5	72.72	75.55	- 2.837
8	10	1.2101	1.1855	5	1	1	4051.6	4221.5	79.07	81.04	- 1.977

(Obs.: observed; calc.: calculated; diff: difference)

Table. 5. Structural parameters of In_2Te_3 Thin Films deposited for various thickness on mica substrates

S.No	Deposited at Temperature(K)	Film Thickness (A^O)	Particle Size (D) nm	Strain (ε) X 10^{-3} lin $^{-2}$ m $^{-4}$	Dislocation density (δ) lin m^{-2}
1	303	800	22.18	1.63	$1.82X10^{15}$
2	303	1600	66.7	0.54	$2.02X10^{14}$
3	353	600	37.9	0.95	$6.25X10^{14}$
4	353	1000	44.41	0.81	$4.55X10^{14}$

The Optical transmission spectra were measured using a double beam UV-VIS-NIR spectrophotometer in the wavelength range from 200 to 2000nm for films deposited on mica substrates. Figure 2 shows the Transmittance (T) with wavelength (λ) for In_2te_3 thin film deposited on mica substrates of various thicknesses. The observed fringe pattern of the transmission spectrum indicates that the films are homogeneous in nature. The interference fringe pattern can be analyzed using the Swanepoel's method [15], which is based on the envelope of the interference maxima and minima, appearing in the spectrum. The method provides basis for the calculation of optical constants with increase of thickness. The intrinsic absorption edge for these films was examined using the relation given by Bardeen et al. [16] which states that the absorption coefficient (α) is related to the incident photon energy(hע) as per the equation,

$$\alpha h\text{ע} = \beta(h\text{ע} - E_g)^x \quad (5)$$

where E_g is the energy band gap, x= ½ for a direct allowed transition and x=2 for non-direct transition. β is the parameter that depends on the transition probability. Figure 3 shows hע(eV) Vs $(\alpha h\text{ע})^2(cm^{-1}eV)^2$ of In_2Te_3 thin film deposited on mica substrate of various thicknesses. A plot of $(\alpha h\text{ע})^2$ against hע was found to be a straight line for the films deposited at different substrate temperatures. The intercepts of these straight line plots on the energy axis at $(\alpha h\text{ע})^2 = 0$ were calculated to represent the band gap energies at the different substrate temperatures. It is observed that the band gap energy calculated by assuming direct transition is found to be in the range 1.112 – 1.146 eV for the films deposited at the temperature 303K and 353K of various thicknesses are in agreement with the few reported data on the optical band gap of In_2Te_3 thin films [17]. In amorphous chalcogenide thin films, the number of defects is higher due to the existence of unsaturated bonds [18]. The increase in the thickness of the films results in a homogeneous network with low density of defects [19] thereby, increasing the optical band gap.

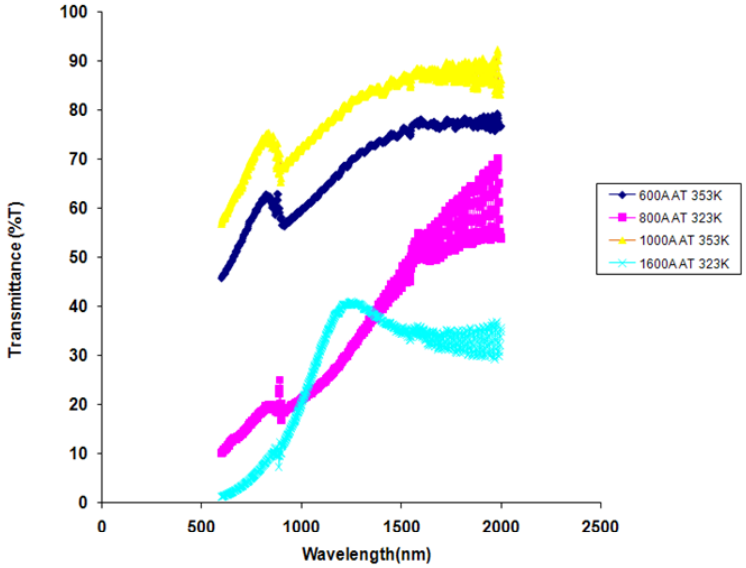


Fig 2. Transmission spectra of In_2te_3 thin films deposited on mica substrate of various thickness

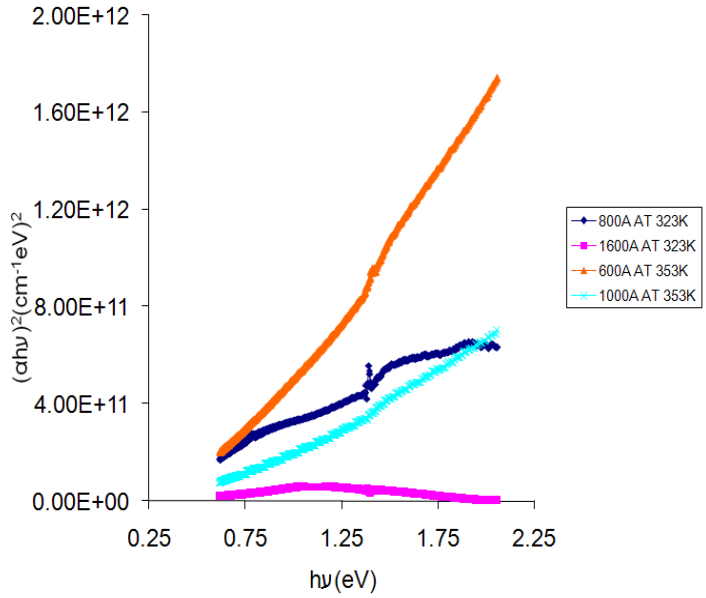


Fig 3. Variation of $(\alpha h\nu)^{1/2}$ versus photon energy ($h\nu$) for different thickness of In_2Te_3 thin films

The refractive index n is deduced from the fringe patterns in the transmittance spectrum. In the transparent region, where the absorption coefficient $\alpha \neq 0$, and the refractive index n of the thin films is given by [15]

$$n = [M + (M^2 - S^2)^{1/2}]^{1/2} \quad (6)$$

where M is given by

$$M = 2S/T_m - (S^2+1)/2 \quad (7)$$

and S is the refractive index of substrate and T_m is minima in the transmittance spectra. In the weak and medium absorption regions, where $\alpha \neq 0$, the refractive index is given by

$$n = [N + (N^2-S^2)^{1/2}]^{1/2} \quad (8)$$

where

$$N = 2s(T_M - T_m)/T_M T_m - (S^2+1)/2 \quad (9)$$

and T_M and T_m are the values of envelope function at the wavelengths in which the transmittance is maxima and minima respectively.

The extinction coefficient k can be calculated from the relation.

$$k = \alpha\lambda/4\pi \quad (10)$$

The variation of refractive index (n) with wavelength of different thickness films of In_2Te_3 thin films is shown in figure 4. Refractive index decreases with increase of wavelength. Variation in refractive index follows the normal dispersion law. Absorption Coefficient (α) and Extinction Coefficient (k) values of In_2Te_3 thin films deposited on mica substrate are reported in Table 6.

The spectral transmittance data for thin film has also been used to derive the complex dielectric function of the thin film. The real and imaginary parts of the dielectric function of the thin film. The real and imaginary parts of the dielectric function, ε' and ε'', are related for n and k [20].

$$\varepsilon' = n^2-k^2 \text{ and } \varepsilon'' = 2nk \quad (1)$$

The dissipation factor is expressed as

$$\tan\delta = \varepsilon''/\varepsilon' \quad (12)$$

The values of Refractive index (n), real (ε') and imaginary (ε") parts of dielectric constant and Dissipation factor (tan δ) deposited on mica substrate are listed in Table 6. It is clearly observed that dissipation factor decreases with the increase in the thickness of films.

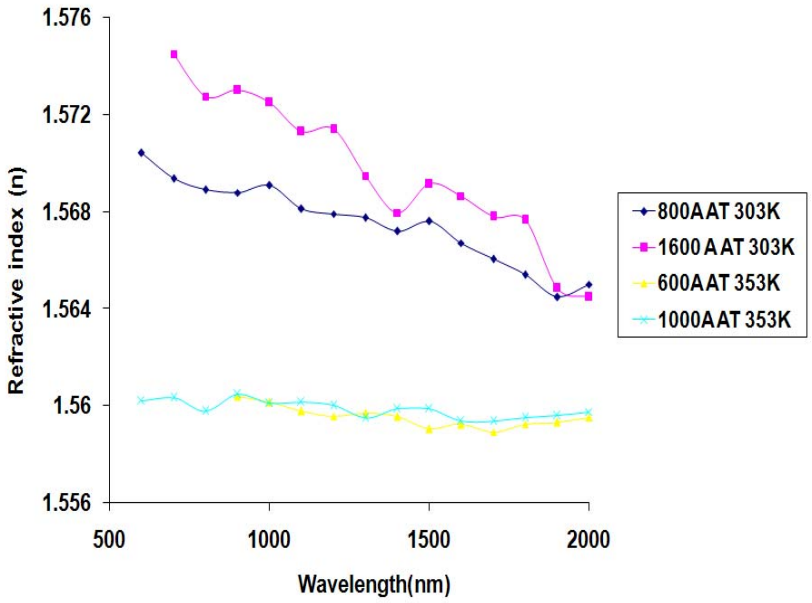


Fig 4. Variation of refractive index (n) with wavelength for different thickness of In_2Te_3 thin films

Table. 7. Absorption Coefficient(α), Extinction Coefficient (k) , Refractive index (n), Real (ε'), Imaginary parts (ε'') of Dielectric constant and Dissipation factor (tan δ) values of In_2Te_3 thin films deposited on mica Substrates

S.No	Film thickness (A^O)	Absorption Coefficient (α) cm^{-1}	Extinction Coefficient (k)	ε' = n^2-k^2	ε ''=2nk	tan δ = ε''/ ε'
1	800	0.66 * 10^6	0.0340	2.4604	0.1066	0.0433
2	1600	0.224 * 10^6	0.0334	2.4732	0.1050	0.0424
3	600	0.724 * 10^6	0.0482	2.4281	0.1502	0.0618
4	1000	0.445 * 10^6	0.3029	2.4339	0.0942	0.0387

4. Conclusions

In_2Te_3 thin films were grown on mica substrate using vacuum evaporation technique with different thickness and substrate temperatures. Increases in optical band gap of the films with the increase of thickness have been attributed to the decrease of localized states in the band gap. The optical constants refractive index(n), Absorption Coefficient (α), Extinction Coefficient (k), Dielectric constant with real(ε') and imaginary(ε''), Dielectric loss (tan δ) and Optical energy gap (E_g) values are calculated from transmission spectra.

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